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White illumination characteristics of ZnS-based phosphor materials excited by InGaN-based Ultraviolet Light-Emitting Diode

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ABSTRACT

White illumination characteristics of ZnS-based phosphor materials exited by an $In_xGa_{1-x}N$-based single quantum well (SQW) -structure ultraviolet (UV) light-emitting diode (LED) have extensively been investigated. In order to evaluate white luminescence, two kinds of ZnS-based white phosphors have been employed. When an UV LED was operated at a current of 10 mA, chromaticity (x, y), color temperature (Tc) and general color rendering index (Ra) of the white luminescence are obtained to be (x, y)=(0.29, 0.33), Tc=7700 K and Ra=70, respectively, for ZnS:Ag + (Zn,Cd)S:Cu,Al phosphors, whilst (x, y)=(0.31, 0.34), Tc=6900 K and Ra=83, respectively, for white phosphor material including ZnS:Cu,Al, Sr and Y materials. The value of chromaticity slightly changed with increasing forward current of the UV light source. As a result, it is possible to obtain stable white luminescence spectrum. The dependence of the luminescence brightness on the thickness of phosphor shows a tendency to saturate for reflection brightness, but for transmission brightness its dependence has a peak due to light scattering effect. The reflection brightness was higher than the transmission brightness. It is revealed that the white luminescence light of stable chromaticity and high brightness using reflection light can be obtained.

Keywords: $In_xGa_{1-x}N$-based UV LED, ZnS-based white phosphor, chromaticity, general color rendering index, reflection brightness, white LED

1. INTRODUCTION

There is now intense interest in the development of high brightness visible and white light-emitting diodes (LEDs) because tremendous technological breakthroughs on the epitaxial growth and LED process of III-V compound semiconductors have been emerged.[1,2,3,4] It is well-known that LEDs can offer advantageous properties such as high brightness, reliability, lower power consumption and long lifetime. The biggest potential application for LEDs will be general lighting.[5] A national program underway in Japan has already suggested that white LEDs deserve to be considered as the general lighting technology of the 21st century owing to an electric power energy consumption.

During the last few years, the technology of white LEDs which is connected with the $In_xGa_{1-x}N$ blue LED coated with a

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YAG:Ce phosphor, has improved the white LED efficiencies from 7.5 lm/W to approximately 30 lm/W. However, the brightness and color purity of InGaN-based white LEDs are dependent on the conditions of phosphor coating, showing "cool white light".

An alternative method of making a color conversion white LED is that a true white can be achieved using three colors from red, green, and blue phosphors excited by ultraviolet (UV) LEDs, similar to three colors based fluorescent lamp. At present, external quantum efficiencies are now as high as 7 % for the UV LED which is operated at about 370 nm. In the future, as the efficiency of blue and UV LEDs will be improved to be approximately 40 %, white LED lamps may replace conventional light bulbs or fluorescent lamps in many applications.

We are for the first time concerned in this paper with the fundamental white illumination characteristics of ZnS-based phosphor materials excited by InGaN-based ultraviolet LEDs. Particularly, two kinds of phosphor materials were used for the characterization of white emission.

2. EXPERIMENTAL PROCEDURES

2.1 Characteristics of an UV LED

Figure1 illustrates the cross section of an $In_xGa_{1-x}N$-based UV LED structure used in the present study. Principally, the LED structure is composed of a single quantum well of $In_xGa_{1-x}N$ whose thickness is estimated to be approximately 50 Å and also of a double heterostructure with a well thickness of 400 Å. The composition of In is nearly zero. The barrier layers are n-and p-type $Al_{0.2}Ga_{0.8}N$. All epitaxial layers were grown on (0001) sapphire substrate by two-flow metalorganic chemical vapor deposition (MOCVD).[4] The typical properties of the UV LED are as follows: an emission wavelength of 370 nm, its linewidth of 11 nm, external quantum efficiencies between 3 and 7.5 % and light output powers between 2 and 5 mW at room temperature (RT).

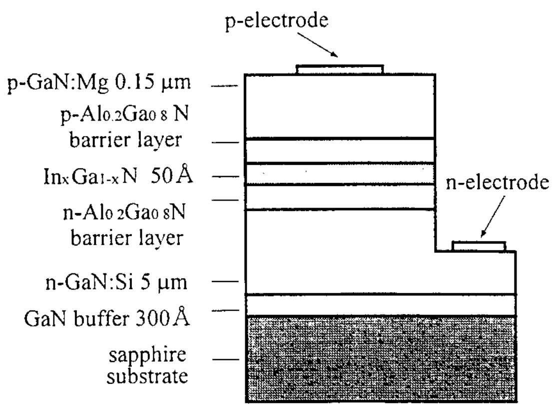

Fig.1 A schematically drawing of the cross section of InGaN/AlGaN SQW UV LED having an active layer of $In_xGa_{1-x}N$ ($x \fallingdotseq 0$).

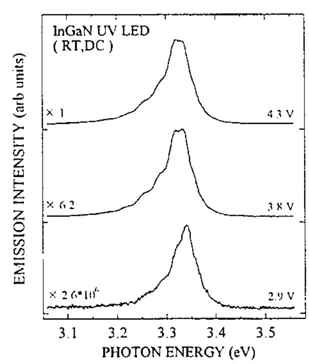

Fig.2 (a) Emission spectra obtained at RT between 2.9 and 4.1 V.

Figure2 (a) shows the electroluminescence spectra obtained at three different forward-biased voltages (2.9, 3.8 and 4.3 V) at RT. The 370 nm emission was observed around 2.9 V and with increasing bias its intensity was increased dramatically up to about 4V(as shown in Fig.2 (b)). Between 2.9 and 4.3 V, the emission peak does not shift with voltage.

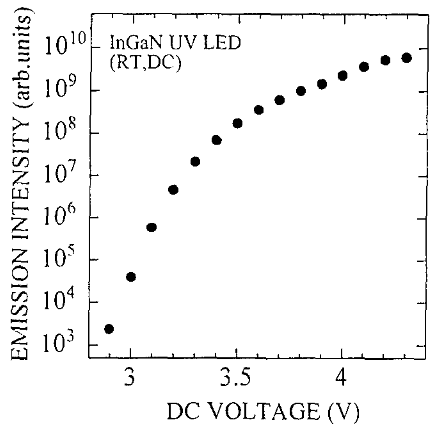

Fig.2 (b) Emission intensity of the 370 nm at RT as a function of forward-bias voltage

2.2 Phosphor materials

Two kinds of phosphor materials based on ZnS were used; ① ZnS:Ag and (Zn,Cd)S:Cu,Al and ②ZnS:Cu,Al including Y- and Sr-components. Excitation spectrum for each phosphor was measured at RT using a CCD camera in conjunction with a 50 cm monochromator under the excitation condition by a Xe lamp (500 W). Excitation bands for each phosphor material were observed in the vicinity of 370 nm at RT.

2.3 Luminescence measurements

Current dependence of luminescence spectra of the phosphors excited by $In_xGa_{1-x}N$ SQW UV LED was measured using CCD camera. Chromatic points in the C.I.E chromaticity diagram and color temperature were evaluated by colorimeter using Si photodiode. Thickness dependence of the luminescence brightness of the phosphors at a current of 10 mA was measured using luminance meter.

3. Three methods for obtaining white lights

Table I shows three methods which can produce white light using LEDs. These technologies are likely to include: 1) RGB LED combinations, 2) binary complementary LED systems and 3) $In_xGa_{1-x}N$-based blue and UV LED systems employing fluorescent phosphors. We need at least three LEDs which primary generate red, green and blue colors from each LED. However, since the different semiconductors are used, one of each primary color must be adjusted by individual supply circuit in order to control the intensity of each color. On the other hand, white LEDs by exciting phosphors have been fabricated using blue and UV LEDs with sufficiently high excitation energy. Using blue LED, the YAG:Ce^{3+} phosphor is excited by 465 nm and then emits yellow fluorescence around 555 nm. The mixture of the blue light from the blue LED chip and the yellow from the phosphor results in a white emission. An alternative method is that a color conversion white LED is also fabricated using both RGB phosphors and UV LEDs.

Table I . Three methods for obtaining white LEDs. Different LED sources, luminescent materials and how to get white light are shown.

Method	LED source	Luminescent materials	Emission mechanism
1 LED chip	Blue LED[6,7,8]	$In_xGa_{1-x}N$/YAG:Ce	Yellow emission from YAG:Ce excited by blue LED
	UV LED[9,10]	$In_xGa_{1-x}N$/R, G, B phosphors	White emission from R, G, B phosphors excited by UV LED
2 LED chips	Blue LED, Blue-green LED, Yellow LED, Amber LED	InGaN, GaP, AlInGaP	W = B + Y =B-G + Amber
3 LED chip	Blue LED, Green LED, Red LED	InGaN, AlInGaP	W = R + G + B

4. EXPERIMENTAL RESULTS AND DISCUSSION

4.1. White LED with ZnS:Ag and (Zn,Cd)S:Cu,Al phosphors

Figure3 shows the dependence of luminescence spectra on forward current: 1, 10 and 20 mA. There appear two emission peaks locating at 453 and 561 nm, respectively. We have obtained chromaticity (x, y), color temperature (Tc) and general color rendering index (Ra) to be (0.29, 0.33), 7700 K and 70, respectively, at a forward current of 10 mA. With increasing forward current, two peaks do not shift at all, but the 453 nm band is slightly increased in intensity.

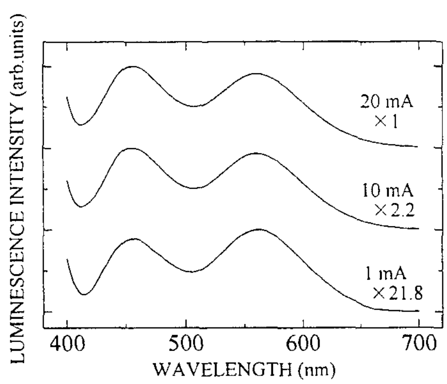

Fig.3 Emission spectra of ZnS:Cu,Al phosphor obtained at RT at three different forward-bias currents (1, 10 and 20 mA)

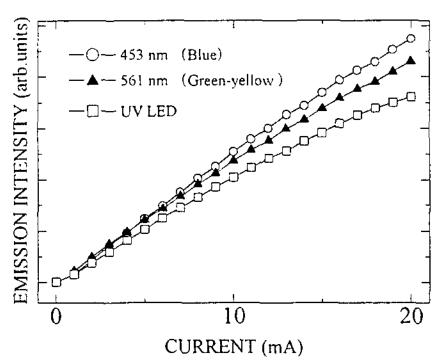

Fig.4 Emission intensities of the 453 nm blue and 561 nm green-yellow emission bands at RT as a function of forward-bias current. The 370 nm UV emission intensities are also plotted against current. The changes in the 453 nm and 561 nm emission intensity with current are normalized.

Figure4 shows the relationship between the 453 nm blue band and 561 nm yellow band intensities, and forward current, where the normalized intensities for each emission band are plotted. This figure also shows the changes in the intensity of the 370 nm UV emission from the UV LED as a function of forward current. Over a forward current of 10 mA the UV emission is saturated in intensity. However, both the 453 nm and 561 nm emission bands are increased in intensities even over 20 mA.

4.2 White LED with RGB phosphors

Figure5 shows the dependence of luminescence spectra on forward current: 1, 10 and 20 mA. There are at least three main peaks locating at 447, 528 and 626 nm, respectively. The 447 nm blue emission, 528 nm green emission and 626 nm red emission bands are originated from fluorescent emission of ZnS:Cu,Al, Sr and Y materials. We have obtained chromaticity, color temperature and general color rendering index (Ra) to be (0.31, 0.34), 6900 K and 83, respectively, at a forward current of 10 mA. Compared to those of the ZnS:Cu,Al phosphor, illumination factors are much improved. The Ra = 83 is close to that of three-band emission fluorescent lamp, indicating the warm white light.

Figure6 (a) shows the relationship between three (RGB) emission band intensities, and forward current, where the normalized intensities for each emission band are plotted. This figure also shows the forward-current dependence of the UV emission band at 370 nm of the UV LED source. Despite the fact that the UV emission is saturated in intensity, three RGB emission intensities are gradually increased up to a current of 20 mA.

The component of red emission is very important to improve the Tc and Ra values. However, the white light with a low Ra is possible to obtain using the blue and green-yellow components. Fig.6 (b) shows the intensity ratio between the blue and green component as a function of forward current of the UV LED. Over a current of 10 mA, the ratio is maintained to be about 1.65, in which the blue component is always higher than that of the green component. Fig.6 (b) also shows the intensity ratio between the red and green component as a function of forward current. Compared to the ratio between the blue and green component, this ratio (2.9-3.0)

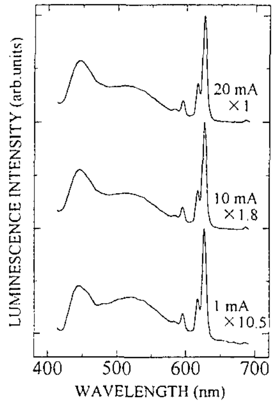

Fig.5 Emission spectra of ZnS-, Y- and Sr-based phosphor obtained at RT at three different forward-bias currents (1, 10 and 20 mA)

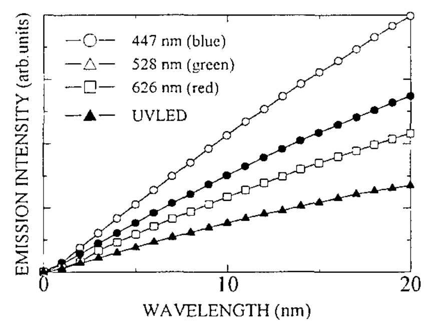

Fig.6 (a) Emission intensities of the 447 nm blue, 528 nm green and 626 nm red emission bands at RT as a function of forward-bias current. The 370 nm UV emission intensities are also plotted against current. The changes in the 447, 528 and 626 nm emission intensity with current are normalized.

of red emission to green emission is almost constant between 10 and 20 mA.

Figure7 represents the forward current dependence of chromatic points of the RGB phosphor between 1 and 20 mA, where similar characteristics of $In_xGa_{1-x}N$ blue LED: YAG phosphor are plotted. As already seen in Figs.5, 6 (a), and 6 (b), the changes in Tc values become small against the increase in the forward current. The use of the RGB phosphors for fabricating white LED is essential to produce stable color temperature and general color rendering index with increasing forward current. On the other hand, YAG phosphor-coated white LED indicates, that the changes in Tc values are decreased up to (x, y) = (0.29, 0.28) in the chromaticity diagram at 20 mA.

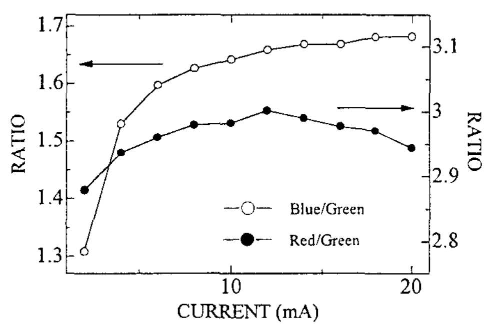

Fig.6 (b) Emission intensity ratios between blue and green component, red and green as a function of forward-bias current

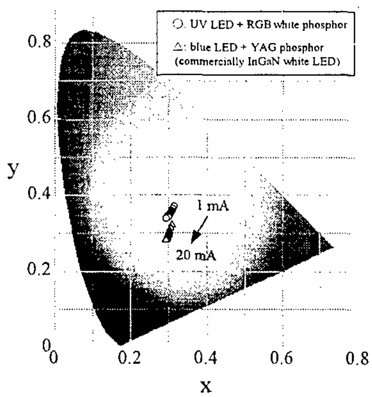

Fig.7 Changes (○) of chromaticity (x,y) coordinates in UV LED excited ZnS-, Y-, Sr-based RGB phosphor between 1 and 20 mA. Similar changes (△) in the blue LED excited YAG phosphor are observed.

4.3 Phosphor thickness dependence of white light intensity under the transmission and reflection conditions

Figures8 show the phosphor layer thickness of white brightness measured under the transmission and reflection conditions for two phosphors materials of ZnS:Ag and (Zn,Cd)S:Cu,Al (a) and ZnS:Cu,Al, Sr and Y system (b), respectively. In both cases, the vertical scale is plotted by luminous intensity. With increasing thickness, the white light intensity shows a tendency to saturate for reflection brightness in Figs. (a) and (b). On the other hand, the white light intensity has a maximum peak for transmission

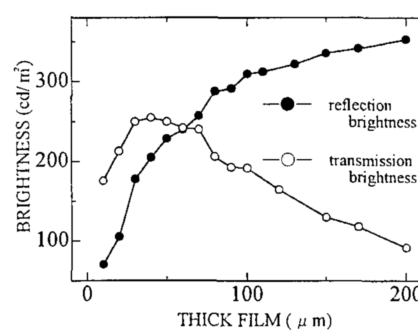

Fig.8 (a) Changes in white light intensity of ZnS:Ag and (Zn,Cd)S:Cu,Al phosphor as a function of phosphor layer thickness under brightness because of the decrease in intensity due to light scattering with thickness. In the case of transmission brightness, the peak is located at about 30~50 μm and at a thickness of 200 μm the transmission intensity is two or three times of magnitude lower than the maximum intensity. On the other hand, in the case of reflection brightness, over the thickness of 100 μm, the intensity becomes saturated and is higher than the transmission maximum intensity.

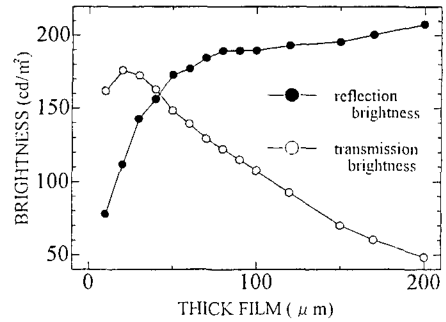

Fig.8 (b) Changes in white light intensity of ZnS-, Y-, Sr-based phosphor as a function of phosphor layer thickness under transmission

5. CONCLUTIONS

We have investigated the white illumination characteristics of two types of ZnS-based phosphor materials excited by an $In_xGa_{1-x}N$-based SQW UV LED. The chromaticity, color temperature and general color rendering index were obtained to be (0.31, 0.34), 6900 K and 83, respectively, in the ZnS-, Y- and Sr-based RGB phosphor. It is noted that the value of chromaticity slightly changes with increasing forward-bias current. The present experimental evidence suggests that the ZnS-based phosphor coating white LED can produce excellent spectral purity with forward current. The dependence of the white brightness on the phosphor thickness shows a tendency to saturate for reflection brightness, whilst for the transmission brightness its dependence has a maximum intensity around the thickness of 30~50 μm. It is indicated that the white light of stable chromaticity and high brightness can be obtained under the reflection condition.

ACKNOWLEDGEMENTS

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