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A Novel Blue Thin Film Electroluminescent Phosphor $Gd_3Ga_5O_{12}:Ag$ *

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A new blue thin film electroluminescence (TFEL) phosphor $Gd_3Ga_5O_{12}:Ag$ is prepared by electron beam evaporation. Photoluminescence and Electroluminescence of $Gd_3Ga_5O_{12}:Ag$ film were investigated. The luminance of TFEL device excited by an alternating-current sinusoidal voltage with frequency of 1000 Hz is about 2 cd/m^2 .

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Alternating-current thin film electroluminescence (ACTFEL) devices are used in the high-resolution flat-panel displays, and thin film electroluminescence (TFEL) technology could provide an attractive solution for flat-panel display once the problem of efficient color phosphor is solved.¹ Today multi-color (red and green) TFEL products have been sold in the display market, developing a bright blue electroluminescence (EL) phosphor is a very important problem for full-color EL displays. $SrS:Ce$ ACTFEL devices² have recently received much attention because of their broad band blue-green light emission, the devices can provide blue emission by filters. But it is more practical to obtain blue emission for full-color TFEL display without using of color filters. $SrGa_2S_4:Ce$ EL devices³ can provide a pure blue emission, but the preparation of $SrGa_2S_4$ is very difficult due to sulfur deficiency etc. So multicomponent oxide host materials may be a good candidate in TFEL. $ZnGa_2O_4$ and $ZnAl_2O_4$ have been investigated in green emission.⁴ Wang *et al.*⁵ have found an efficient blue photoluminescence composite material $Gd_3Ga_5O_{12}/SiO_2$. In this letter, We report on a blue emitting TFEL of a new phosphor $Gd_3Ga_5O_{12}:Ag$.

$Gd_3Ga_5O_{12}:Ag$ was prepared by mixing three powders: Gd_2O_3 (purity 99.99%), Ga_2O_3 (99.99%) and $AgNO_3$ (99.9%). The portions of the mixture were chosen in order to get a stoichiometric material with about 5 mol% Ag^+ (the luminescent center). The mixture was pressed into pellets, then they were sintered as follows: 200°C for 150 h, 400°C for 24 h, 600°C for 12 h, 1000°C in Ar atmosphere for 2 h.⁵ The structure of our EL devices was ITO/SiO/SiO₂/phosphor/SiO₂/SiO/Al (Fig. 1). The thickness of SiO, SiO₂, and $Gd_3Ga_5O_{12}:Ag$ were 40, 25, and 150 nm, respectively, and they were deposited on the quartz substrate with ITO by electron beam evaporation at nominal deposition rate 0.3, 0.13, and 0.04 nm/s, respectively. In the process the substrate temperature was kept at 275°C. The samples were annealed in Ar at 600°C for 4 h.

The crystallinity of the phosphor powder and thin films was measured by x-ray diffraction with Rigaku Rint-2400 diffractometer using $Cu K\alpha$ radiation and graphite monochromator. The spectra of photoluminescence (PL) and EL for the thin film phosphor was measured by HITACHI-4010 Fluorescence Spectrophotometer. In the measurements, the TFEL devices were excited by an applied voltage with 1000 Hz frequency.

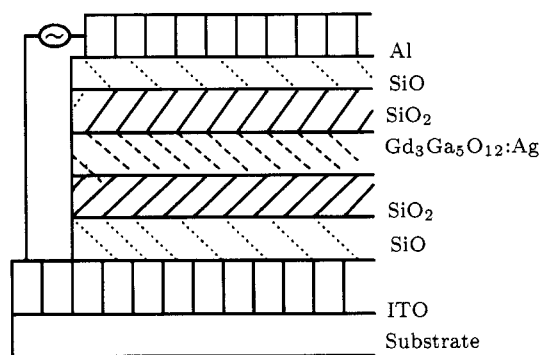


Fig. 1. Structure of the devices.

Figure 2 shows the diffraction patterns of phosphor powder, annealed and as-deposited thin films. The diffraction pattern for the as-deposited films shows a broad band indicating that unannealed films are amorphous. The diffraction peak positions for annealed films closely match those of the powder from 25° (value of 2θ) to 40°. The ratio of the diffraction peak intensity at $2\theta = 28.9^\circ$ (reflection at 400 orientation) to that at $2\theta = 32.3^\circ$ (reflection at 420 orientation) in annealed films is much higher than that measured for the powder sample. Annealed films thus have moderately preferred (400) orientation assuming that in the powder the crystal grain has random orientation. The full width of half maximum (FWHM) of diffraction peak for the (400) reflection from the annealed film is circa 0.4° , that shows a good crystallinity of thin film.

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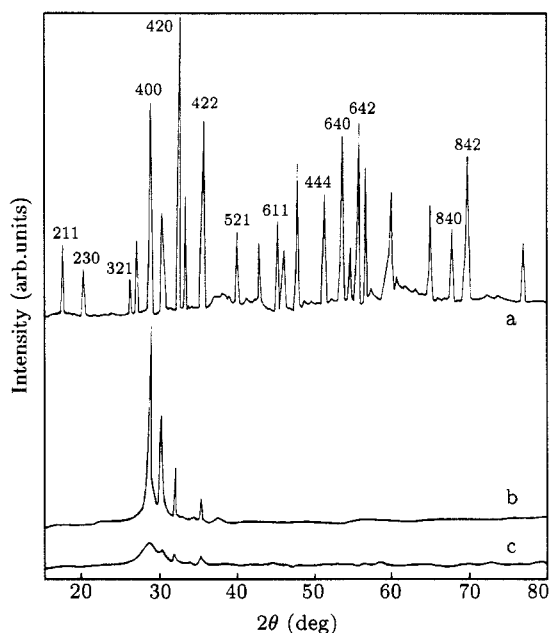


Fig. 2. XRD patterns of $\text{Gd}_3\text{Ga}_5\text{O}_{12}:\text{Ag}$ powder (a) and thin films, both annealed (b) and as-deposited (c).

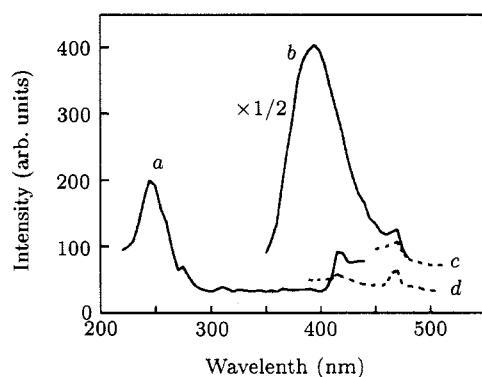


Fig. 3. PL excitation spectrum (a), emission spectrum excited by beams at 245 nm (b), 275 nm (c), and 416 nm (d) of an annealed $\text{Gd}_3\text{Ga}_5\text{O}_{12}:\text{Ag}$ (5 mol%) film.

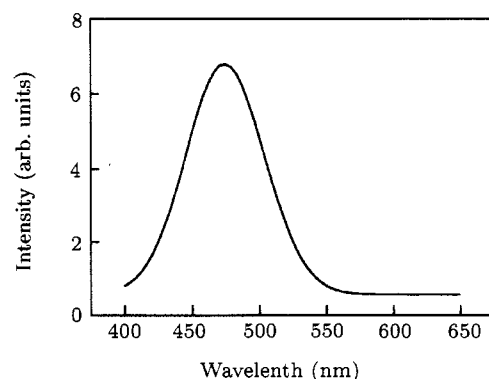


Fig. 4. EL spectrum of $\text{Gd}_3\text{Ga}_5\text{O}_{12}:\text{Ag}$ thin film.

In the PL, three peaks (Fig. 3) at 245, 275, and 416 nm can be observed in excitation spectrum for the monitor wavelength of 469 nm. The intensity of 245 nm is much higher than those at 275 and 416 nm.

Figure 3 also shows the PL emission of the phosphor excited by beams at 245, 275, and 416 nm, respectively. The PL excited by 245 nm has two peaks located at 397 and 469 nm, and PL spectra excited by beams at 275 and 416 nm have only one peak located at 469 nm. The peak at 245 nm is thought to be due to the inter-band absorption of phosphor, and those at 275 and 416 nm due to the transitions from the ground state of Ag^+ to the excited state of Ag^+ . We have also prepared a series of devices with phosphor containing different concentrations of Ag^+ (0.1–8 mol%), the intensity of PL at 469 nm increases with the concentration of Ag^+ . When the concentration of Ag^+ increases up to 0.5 mol%, the intensity of PL at 469 nm is saturated. So we conclude that the peak at 469 nm of PL originates from the transitions from the excited state to the ground state of Ag^+ . The only PL excitation spectrum is located at 245 nm when the monitor wavelength is 397 nm (not shown in the illustration). The emission at 397 nm is considered to be the transitions from conduction band to the ground state of Ag^+ .

Figure 4 shows the EL spectrum by the direct impact excitation. The peak of EL emission located at 470 nm is thought to be originated from the excited state ($4d^95s$) to the ground state ($4d^{10}$) of Ag^+ . While we do not find the EL emission at 397 nm corresponding to that of PL, it means that the energy of the hot electrons is not large enough to excite the electrons from ground state to the corresponding excited levels. The luminance of these devices is about 2 cd/m^2 at 30 V above the threshold, when frequency of the sinusoidal voltage is 1000 Hz. The conditions for higher energy of hot electrons and higher luminance are to be investigated later.

In conclusion, we have prepared a new blue TFEL phosphor $\text{Gd}_3\text{Ga}_5\text{O}_{12}:\text{Ag}$, the phosphor has a good crystallinity after the post annealing at 600°C for 4 h. The PL spectrum has two peaks: 397 and 469 nm, and the EL spectrum is pure blue at 470 nm. The luminance of the TFEL devices is about 2 cd/m^2 when an excitation voltage of 30 V above the threshold at a sinusoidal wave frequency 1000 Hz is applied. We think that this phosphor will be a good candidate for film electroluminescence.

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