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*Research Article*

# Low-Cost ZnO:YAG-Based Metal-Insulator-Semiconductor White Light-Emitting Diodes with Various Insulators

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ZnO:YAG-based metal-insulator-semiconductor (MIS) diodes with various insulators were synthesized on an indium tin oxide (ITO) glass by ultrasonic spray pyrolysis. $SiO_2$ and MnZnO (MZO) were separately used as insulators. X-ray diffraction revealed the crystalline structure of the ZnO:YAG film. The photoluminescence (PL) properties of the ZnO:YAG film were studied and the color of photoluminescence was found to be almost white. The electrical properties of the diodes with different insulators and thicknesses were compared. The diode with the $SiO_2$ insulator had a lower threshold voltage, smaller leakage current, and a higher series resistance than that with the MZO insulator layer.

## 1. Introduction

Zinc oxide (ZnO) is II–VI compound semiconductor with a wide direct band gap (3.36 eV); it has a large exciton binding energy of 60 mV and a hexagonal wurtzite structure. These excellent physical properties and easy, low-cost synthesis make ZnO a promising material to replace III-nitride semiconductors for short-wavelength optoelectronic applications, such as blue/ultraviolet (UV) light emitting diodes (LED) [1–5]. Today, the most common white LED is fabricated from blue LEDs that are made of InGaN and coated with phosphors of different colors. ZnO is easier and cheaper to fabricate than the InGaN-based LED device. ZnO can be conveniently deposited over a large area for advanced lighting applications. Numerous ZnO nanorod-based or ZnO nanotube-based white LEDs have been developed in the past few years [6–9]. Most of the aforementioned devices require p-GaN to form a heterostructure and have a complex fabrication process. However, we have already demonstrated that the photoluminescence of ZnO:YAG is almost white [10]. Therefore, the ZnO:YAG film can be used to fabricate LEDs that emit white light using a simple ultrasonic spray process.

In this work, a ZnO:YAG-based MIS white LED is developed. $SiO_2$ and MnZnO (MZO) were used as insulators. The effects of various thicknesses of insulators were compared. The electrical properties and crystallinity of the ZnO:YAG film were examined by making Hall measurement and by X-ray diffraction (XRD) analysis. The PL measurements were carried out to study the luminescence of the prepared devices.

## 2. Experimental Details

Figure 1 schematically depicts the structure of the ZnO:YAG-based MIS LED. The ZnO:YAG layer with a thickness of 1 $\mu$m was deposited on a commercially available ITO/glass substrate by ultrasonic spray pyrolysis [10]. An aerosol of the precursor solution, which consisted of zinc acetate, ammonium acetate, and YAG phosphor (at 1 wt.%; NYAG4156 phosphor, INTEMATIX, Fremont, CA, USA) powder, was produced using a commercial ultrasonic nebulizer. Then, $SiO_2$ insulators with various thicknesses and ITO electrodes were deposited on the as-prepared ZnO:YAG films by RF sputtering for comparison. Table 1 presents the flow rate of argon, substrate temperature, sputtering power, and chamber pressure during the deposition by sputtering. The MZO insulator layer was also deposited by ultrasonic spray pyrolysis with a precursor solution that consisted of zinc acetate, ammonium acetate, and manganese chloride [11]. The

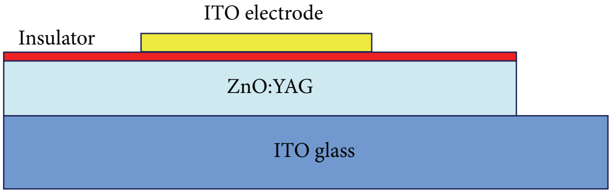
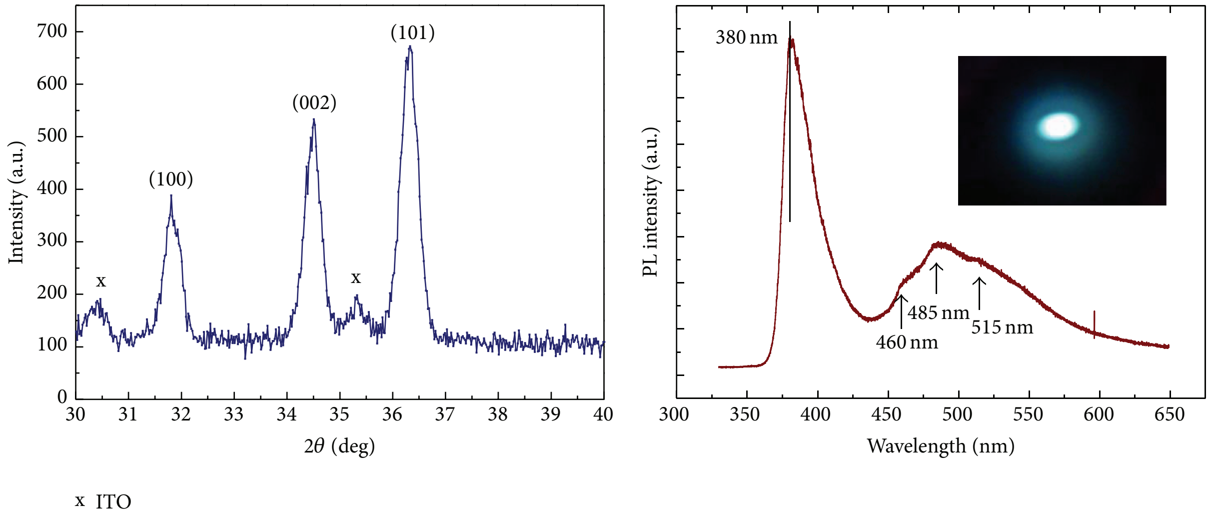


Figure 1: ITO/insulator/ZnO:YAG MIS structure on ITO/glass substrate.



(a) (b)

Figure 2: (a) XRD pattern and (b) RT PL spectrum of ZnO:YAG film on ITO/glass substrate deposited by ultrasonic spray pyrolysis. Inset shows photograph of photoexcited luminescence.

Table 1: Parameters of deposition of insulator and ITO electrode layers by sputtering.

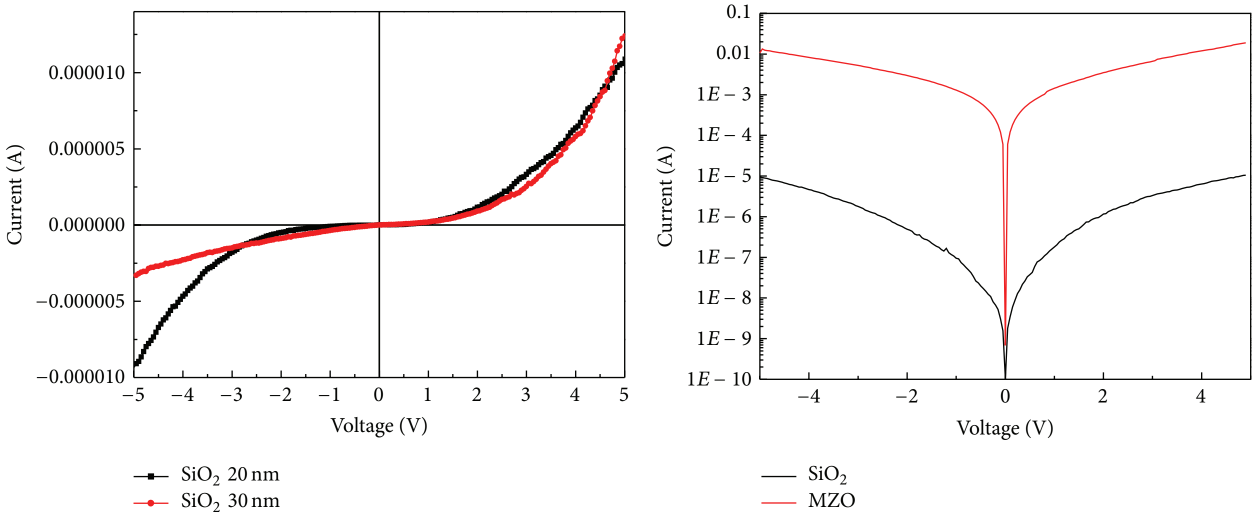
| | Flow rate of argon (sccm) | Substrate temperature (°C) | Sputtering power (W) | Chamber pressure (mTorr) |
|---|---|---|---|---|
| $SiO_2$ | 40 | RT | 60 | 4.23 |
| ITO electrode | 40 | RT | 50 | 3 |

Hall measurements revealed that ZnO:YAG exhibits n-type conduction with an electron concentration of approximately $10^{18}$ $cm^{-3}$. The current-voltage ($I$-$V$) characteristics of the devices were measured using a Keithley 2400 electrometer. The crystalline microstructure of the ZnO:YAG film was determined by X-ray diffraction with Cu-K$\alpha$ radiation ($\lambda$ = 0.1541 nm) in the scanning range of $2\theta$ from 30° to 40°. The PL spectrum was obtained by a Dongwoo spectrophotometer (Dongwoo, Soule, Korea) at room temperature by exciting the ZnO:YAG using an He-Cd laser (325 nm).

## 3. Results and Discussion

Figure 2(a) shows a typical XRD pattern of the ZnO:YAG film that was deposited on an ITO/glass substrate that was prepared by the ultrasonic spray pyrolysis. The spectrum includes broad peaks at positions 31.82°, 34.52°, and 36.34°, which are strongly associated with the (100), (002), and (101) planes of the ZnO phase. This finding suggests that the thin film was polycrystalline and has a ZnO phase with a hexagonal wurtzite structure (by JCPDF no. 75-0576).

Figure 2(b) displays the room-temperature photoluminescence (RT PL) spectrumof the ZnO:YAG film. The dominant peak at 3.26 eV (380 nm) corresponds to the optical band gap of ZnO films with a wide band gap and can be attributed to the recombination of free excitons in an exciton-exciton collision process [12, 13]. The visible luminescence, emitted over a wide range from 450 nm to 600 nm, is composed of at least three broad peaks. The peaks at 460 nm, 480 nm, and 515 nm are attributed to electron transfer from the zinc interstitial level ($Z_{ni}$) to the oxygen vacancy ($V_o$) defect level, which may be caused by the incorporation of YAG phosphor [14]. The broad peak in the visible range may also include a peak at 540 nm, which is associated with emission by the YAG phosphor. The photoexcited luminescence of the ZnO:YAG film is almost white, as can be seen in the inset photograph in Figure 2(b). The 380 nm UV emission and the wide visible emission band ranging from 450 to 600 nm contribute together to the white light, as mentioned above such that the color of electroluminescence (EL) from the ZnO:YAG-based MIS LED should be white.



(a)

(b)

Figure 3: (a) $I$-$V$ characteristics of ZnO:YAG-based MIS LED with SiO$_2$ insulator layer of varying thickness. (b) $I$-$V$ characteristics of ZnO:YAG-based MIS LED with different insulator layers with a thickness of 200 nm.

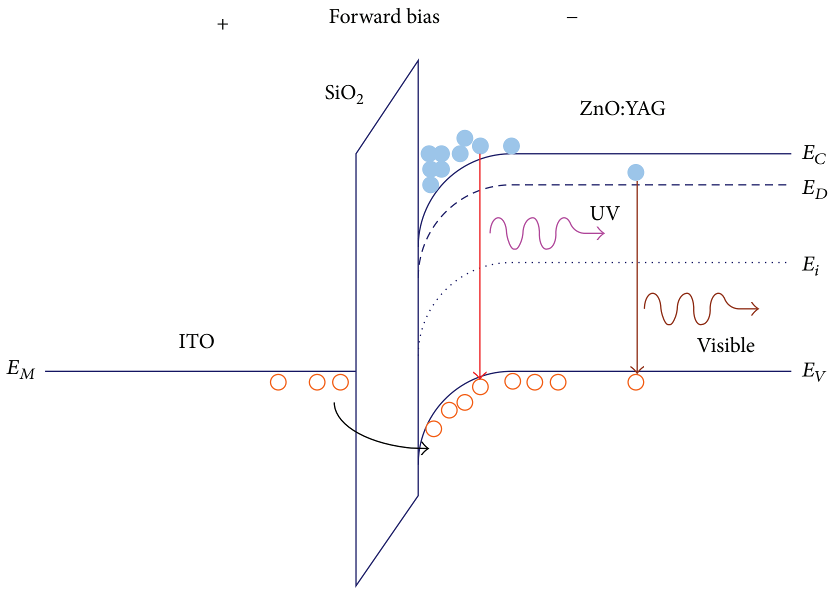


Figure 4: Energy band diagram of ZnO:YAG-based MIS LED with SiO$_2$ insulator layer under forward bias.

Figure 3 plots the $I$-$V$ characteristics of the ZnO:YAG-based MIS LED with different insulator layers of various thicknesses. The device size is 3 mm × 3 mm. As presented in Figure 3(a), the $I$-$V$ curves of the devices with SiO$_2$ insulator layers exhibit diode-like rectifying behavior to some extent. The forward threshold voltage is ~3 V. Figure 3(b) reveals that the ZnO:YAG-based MIS LED with the SiO$_2$ insulator layer has a lower leakage current and a higher series resistance than that with the MZO insulator layer. The forward threshold voltage of the device with the MZO insulator layer is ~2 V.

Figure 4 presents the energy band diagram of the ZnO:YAG-based MIS LED with an SiO$_2$ insulator layer under forward bias. The energy band of ZnO:YAG under the SiO$_2$ layer is bent downward under forward bias. Therefore, tunnel injection of holes from the ITO substrate into the valence band of ZnO:YAG film via surface states in the insulator layer occurs. The SiO$_2$ energy barrier is so large leading to induce an accumulation layer of electrons at the SiO$_2$/ZnO:YAG interface. Many of the holes that are injected from the ITO substrate are recombined with the electrons confined in the downward-bending region of the conduction band of ZnO:YAG film. The confined electrons and the defect levels ($E_D$) of ZnO:YAG film recombine radiatively with the injection holes in the valence band and then generate UV and visible emission, respectively. The white emission from the ZnO:YAG-based MIS LED can be theoretically understood as being generated by blending UV and visible emission with YAG photoexcited emission.

## 4. Conclusion

The ZnO:YAG film herein has the hexagonal wurtzite structure and emits PL that is almost white. ZnO:YAG-based MIS LEDs with different insulator layers were successfully prepared using a low-cost, simple, but effective ultrasonic spray pyrolysis method. The diode with the $SiO_2$ insulator had a lower threshold voltage, smaller leakage current, and a higher series resistance than the one with the MNO insulator layer. The modal of the energy band of the device has been also addressed. The white emission from the ZnO:YAG-based MIS LED can be theoretically understood as being generated by blending UV and visible emission with YAG photoexcited emission. The study implies that the ZnO:YAG film is a promising material for fabricating white LED with low cost.

## Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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