Properties of ZnO:Ga Thin Films Deposited by dc Magnetron Sputtering: Influence of Ga-Doped Concentrations on Structural and Optical Properties

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Properties of ZnO:Ga Thin Films Deposited by dc Magnetron Sputtering: Influence of Ga-Doped Concentrations on Structural and Optical Properties

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Abstract: ZnO:Ga thin films were deposited on corning glass by dc magnetron sputtering. Influence of Ga-doped concentrations on the structural and optical properties of ZnO:Ga thin films were investigated. The XRD patterns show that the crystallinity of deposited films improved with the increase of Ga concentrations from 1 to 2%, then decrease at 3% Ga concentrations. The optical transmittance of films with 1% and 2% Ga concentration reach 85% in the visible range, while at 3% Ga concentration the transmittance of film only 70%. We observed that the band gap of film change due to the addition of Ga dopant. The band gap of the films are 3.27, 3.28 and 3.21 eV for 1, 2 and 3% Ga-doped concentrations, respectively.

Keywords: ZnO:Ga Thin Films, DC Magnetron Sputtering, Structural, Optical Properties

Introduction

Transparent conductive oxide (TCO) such as indium tin oxide (ITO) thin films has been widely used as transparent electrodes, window materials for display and solar cells (Kao *et al*., 2012). Many studies have been conducted in order to obtain an alternative material of ITO for TCO application. This is because although possess fascinating properties, the price of ITO is expensive. ZnO is a promising alternative material for ITO in the TCO applications due to possess some promising properties such as inexpensive, non-toxic, low deposition temperature and chemically stabile (Ma *et al*., 2007) as well as wide optical band-gap (3.4 eV) (Lee, 2013). Nevertheless, the transmittance and electrical conductivity of ZnO material is inferior to ITO. Besides, the properties of the pure ZnO are unstable (Yang *et al*., 2009). Therefore, further treatment for upgrading the properties of ZnO is required. The enhancement of properties could be done by precisely control the growth processes and use several appropriate dopants (Nayeef *et al*., 2013).

Shin *et al*. (2009) have deposited ZnO films by introducing B, Al, Ga and In dopants. They have reported that the atom of dopants replace the Zn site in the ZnO crystal that one more free of electron was generated. From all dopant elements, Ga is the most effective for ZnO (Yang *et al*., 2009). Ga elements were selected due to the length of Ga-O bond (1.92 Å) almost corresponds to Zn-O bond (1.97 Å). As a result, the lattice mismatch with ZnO is very small. So the addition of Ga doping in the some extent is expected increase the transmittance and conductivity of films but does not damage the crystal structure of ZnO as host material. It because the crystal structure is damaged, the transmittance and the conductivity of films would decrease.

Numerous studies have reported the deposition of ZnO:Ga films with variation deposition techniques such as atomic layer deposition (Maeng and Park, 2013), sol-gel method (Lin *et al*., 2010), chemical vapour deposition (Yang *et al*., 2009), physical vapour deposition (Lee, 2013), pulsed laser deposition (Shin *et al*., 2009), magnetron sputtering (Sheu *et al*., 2007), DC reactive magnetron sputtering (Ma *et al*., 2007) and RF sputtering (Yu *et al*., 2005). It has been reported that the properties of ZnO:Ga thin films are dependent on the deposition methods.

Among this techniques sputtering method possess several advantages such as the films can be deposited in large area of substrate with relatively high of growth rate

(Zhang *et al*., 2002). Besides, sputtering method is considered as a technique with cost-effective in the use of source materials (Ma *et al*., 2007). The source material in the pellet form can be used repeatedly, while the resulted films have high similarity in the quality (Marwoto *et al*., 2016).

The objective of this study is to investigate the influence of Ga-doped concentrations on the structural and optical properties ZnO:Ga thin films grown by dc magnetron sputtering. The structural parameters of films such as the lattice constant, crystal size, *d*-spacing, lattice strain and lattice stress were determined based on XRD results, while the percentage of transmittance and the optical band-gap were determined based on UV-vis analysis. The growth parameters that used in this study is the optimum parameters of ZnO films deposition that had previously been conducted (Marwoto *et al*., 2014).

Materials and Methods

A homemade dc magnetron sputtering system (Marwoto *et al*., 2016) was used to growth ZnO:Ga thin films on corning glass at 400°C of substrate temperature. A sintered target with a mixture of ZnO (99.999%) and Ga_2O_3 (99.999%) was used as the source materials. The target diameter was 2.5 cm with total mass of 10 g. The amount of Ga_2O_3 that added to the target was varied at concentration of 1, 2 and 3 (wt.%). The dc sputtering power and deposition time were kept constant at 30 watt and 1 h, respectively. Corning glass substrates were cleaned using acetone and methanol solution for 15 minutes in the ultrasonic bath.

The crystallographic properties of the deposited films were analyzed using X-Ray Diffraction spectroscopy (XRD) with $Cu-K_{\alpha}$ radiation (1.5406 Å). The optical transmittance measurement was conducted by UV-Vis spectroscopy. All samples were characterized at room temperature.

Results and Discussion

Figure 1 shows the XRD spectrum of ZnO:Ga thin films at three different Ga-doped concentration (wt%). As shown in Fig. 1, a strong (002) peak and a weak (004) peak are observed for all samples. The crystalline dimension of 2θ along *c*-axis is 34.57, 34.44 and 34.05 for 1, 2 and 3 % Ga concentration, respectively. These peaks indicate that the crystal structure of films are independent to the Ga concentration. However, the film deposited with 3% Ga concentration showed a weak (101) peak. It indicated that all of the deposited films are polycrystalline with wurtzite structure and had a preferred orientation with *c*-axis that perpendicular to the substrates (Yu *et al*., 2005). We found that Ga_2O_3 phase was not found on the XRD patterns of deposited films. It indicates that Ga atoms are likely substitute Zn atoms in the hexagonal lattice or segregate to the non-crystalline region to form Ga-O bond. Ma *et al*. (2007) have reported that Ga atoms are able to ionize into Ga^{3+} then substitute Zn^{2+} and generate a free electron from each Ga atoms.

These XRD patterns also showed that the increase of Ga content is change the diffractions intensity. As the concentration of Ga doped increases from 1% to 2%, the (002) peak intensity is increased. When the concentration of Ga is increased to 3%, the intensity of the (002) peak is decreased, but the (001) peak is observed. The (001) peak was not observed on the ZnO:Ga film with 1% and 2% consentration of Ga. The lowering of (002) peak intensity and the existing of (001) peak indicated the occurring structure degradation of films. This indicated that over quantity of Ga atoms in the ZnO crystal configuration leads to the decrease of crystalinity (Lin *et al*., 2010). Although, the existing of Ga atoms in the ZnO crystal could improve the crystallinity, but if the Ga amounts are over quantity, it leads to the defect of the crystal structure. The increase of Ga concentrations in the ZnO crystal would increase the repulsive force arising from the existing positive charges from the Ga^{3+}, then generate the slight lattice deformation and disorder in the ZnO crystal as host material (Yang *et al*., 2009). This result is similar to the Maeng and Park (2013) work. They reported that the (002) peak intensity of ZnO:Ga films prepared by atomic layer deposition is significantly decreased with the increase of Ga-doped concentration.

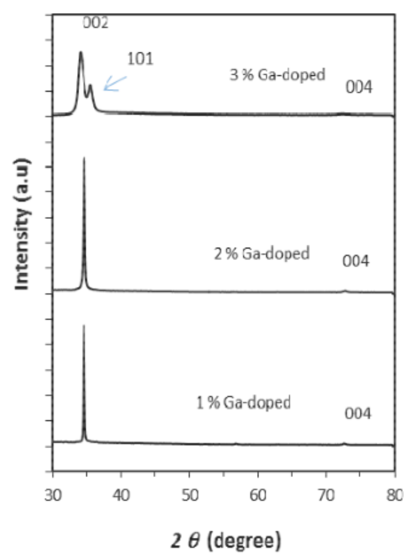

Fig. 1. X-ray diffraction patterns for ZnO:Ga thin films deposited at different Ga-doped concentration

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Table 1 shows structural parameters of ZnO:Ga thin films. The crystallite size of ZnO:Ga films have been determined from the FWHM of diffraction peak using the Scherrer formula as expressed in Equation 1.

$$D = (0.9\lambda) / (\beta \cos\theta) \quad (1)$$

where, D is the diameter of the crytallites film, λ is the wavelength of $Cu-K_{\alpha}$ (1.5406 Å), β is the FWHM and θ is the Bragg angle.

From the Table 1, it can be seen that the Ga-doped has a significant contribution on the change of the crystal size of deposed films. The crystal size decreases from 42 to 12 nm with the increment of Ga concentration, while the lattice constant of *c*-axis increase from 0.519 to 0.526 Å. The enhancement of the *c*-axis lattice is considered due to the increment of Ga^{3+} ions that substitute of Zn^{2+}, then increase the total repulsive force as previously explained.

Table 2 shows the lattice strain and stress of ZnO:Ga thin films with different Ga-doped concentration. The lattice strain and stress were obtained from the XRD spectra analyzed. The lattice strain can be calculated by using Equation 2 (Maeng and Park, 2013).

$$\varepsilon = \frac{\beta}{4\tan\theta} \quad (2)$$

where, ε refers to lattice strain, β is the full width half maximum (FWHM) and θ is diffraction angle. The Stress of films is given by Equation 3.

$$\sigma_{film}^{XRD} = -233\varepsilon \quad (3)$$

where, σ_{film}^{XRD} denotes the stress of thin film.

Figure 2 shows the transmittance spectra of ZnO:Ga thin films deposited with various Ga-doped concentration. It can be seen that the transmittance of films with 1% and 2% of Ga-doped concentration achieve more than 80% in the visible region. The transmittance of films enhance with the increasing of Ga concentration. This transmittance value is similar with the transmittance spectra of the ITO thin films that deposited by Shin *et al.* (2009). Meanwhile, the transmittance of ZnO:Ga film with 3% Ga-doped concentration only reach 70%. This result is strongly agree with the structural properties of films i.e. the ZnO:Ga(3%) film has lowest degree of crystallinity hence it is not surprising perform with the lowest transmittance. The transmittance is strongly affected by the crystallinity of the films (Lee, 2013; Li *et al.*, 2013).

ZnO:Ga thin films have a direct band gap, so that the absorption edge of inter band transition is given by Equation 4:

$$(\alpha h\nu)^2 = A(h\nu - E_g) \quad (4)$$

where α refers to the absorption coefficient and A denotes the constant for a direct transition. The band gap energy (E_g) of film is obtained by plotting α^2 vs. $h\nu$ and extrapolating the straight line of this plot to the energy (axis) as shown in Fig. 3.

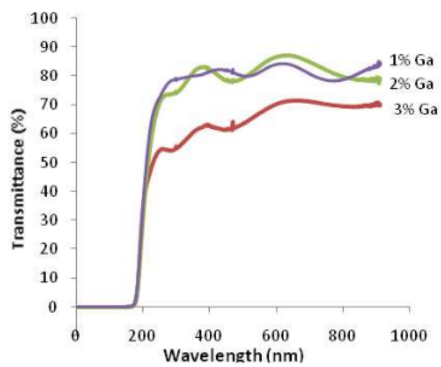

Fig. 2. The transmittance spectra of ZnO:Ga thin films deposited at different Ga-doped concentration.

Table 1. Structural parameters of gallium doped zinc oxide thin films deposited at different Ga-doped concentration

Ga-doped (%)	2θ (°)	Lattice constant *c* (Å)	Crystal size (nm)	d-spacing (Å)
1	34.57	0.519	42	2.59
2	34.44	0.520	24	2.60
3	34.05	0.526	12	2.63

Table 2. Lattice strain and stress value of gallium doped zinc oxide thin films deposited at different Ga-doped concentration

Ga-doped (%)	Lattice strain	Stress (GPa)
1	0.1555	-36.2234
2	0.2068	-48.2022
3	0.5791	-134.9198

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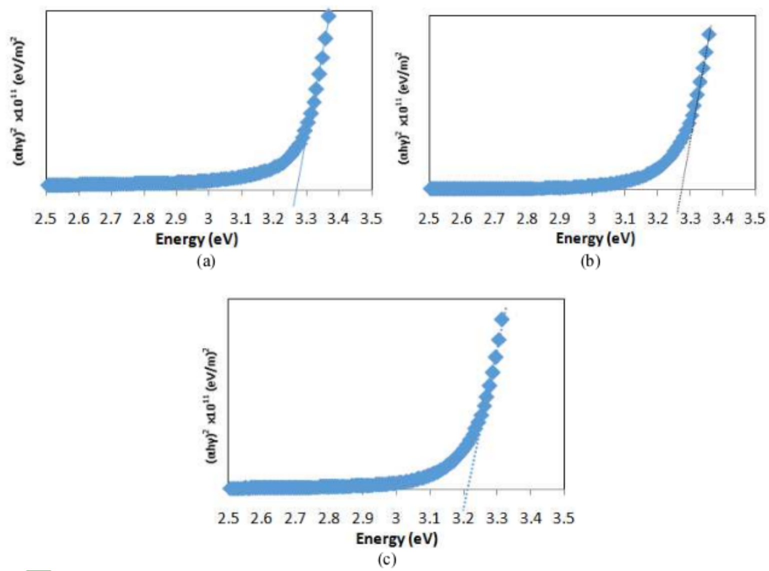

Fig. 3. The band gap (E_g) of ZnO:Ga thin films with different Ga-doped concentration: (a) 1%; (b) 2%; and (c) 3%.

Table 3 shows the band gap of ZnO:Ga thin films deposited at 1, 2 and 3% Ga-doped concentrations. When the Ga concentration increases from 1 to 2%, the band gap, E_g increase from 3.27 to 3.28 eV, even though the increment of the band gap is not significant, while the band gap of the ZnO:Ga (3%) film is 3.21 eV. Thus, it is showed that the width of band gap affect the transmittance. Maeng and Park (2013) have deposited ZnO:Ga thin films by atomic layer deposition. They reported that the band gap of ZnO:Ga thin films with 1-5% Ga-doped concentrations are 3.27 to 3.60 eV. It can be seen, although we use home-made dc magnetron sputtering for deposition process, we obtained the deposited films with comparable band gab to the ZnO:Ga films that deposited by atomic layer deposition method. This phenomenon confirms that the dc magnetron sputtering is a promising method for deposition ZnO:Ga films

Based on Table 3, it can be seen that the band gap of films increase from 3.21 to 3.28 eV as the Ga concentration is decreased from 3 to 2%. The widening of the optical band gap can be explained by Eq. 5 (Shin *et al.* 2009).

$$\Delta E_g = \frac{\hbar^2}{8m^*}\left(\frac{3}{\pi}\right)^{2/3} n_e^{2/3} \quad (5)$$

where, ΔE_g denotes the shift in the doped semiconductor as compared to the un-doped ZnO; m^* is the electron effective mass in the conduction band; $\hbar$ is the Planck'sconstant; and n_e is the electron carrier concentration.

Table 3. Optical band gap of gallium doped zinc oxide thin films deposited at different Ga-doped concentration

Ga-doped (%)	Band gap (eV)
1	3.27
2	3.28
3	3.21

Indeed, the shortening of the optical band gap is an indicator of Burstein-Moss (BM) effect. The BM effect occurs when the Fermi level enters into the conduction band (Li *et al*., 2013). Furthermore, the shrinking of band gap is likely due to the carrier concentration effect (Ma *et al*., 2007). In previous work, Shin *et al*., (2009) reported that the optical band gap would decrease with decreasing of carrier concentration. Maeng and Park (2013) reported that over 5% of Ga doping, the carrier concentration of ZnO:Ga films decrease with the increasing of doping concentrations. On the other hand, Lin *et al*. (2010) also reported that the carrier mobility dropped when Ga doping was laid between 3 to 10 %.

The correlation of optical band gaps widening with the carrier concentration has been reported by Shin *et al*. (2009) and Ma *et al*. (2007). They reported that the optical band gap increase with the increasing of carrier concentration. Thus, the carrier concentration increases as Ga-doped increases from 1 to 2%, while the carrier concentration decreases as Ga-doped increases from 2 to 3%. The increment of carrier concentration was considered due to the improvement of crystallinity (Yu *et al*., 2005). In this case, the doping of Ga atoms replace

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the Zn^{+} sites in the crystal. This phenomenon causes the increase of the carrier concentration (Shin *et al*., 2009). On the other hand, the decreasing of carrier mobility could reduce film conductivity (Lin *et al*., 2010). Yu *et al*., (2005) have reported that ZnO:Ga thin films doped by 3% Ga concentrations also produce film with low electrical conductivity i.e. 1.08×10^{3} $(\text{ohm cm})^{-1}$ at room temperature. Generally, however, the doping of group-III atoms such as Al, In and Ga leads to the increment of electrical conductivity of ZnO:X thin films (X can be Al, In or Ga) compared with un-doped ZnO thin films (Shin *et al*., 2009).

The ZnO:Ga thin films have Zn-O and Ga-O covalent bonds. The lengths of Ga-O bond and Zn-O bond are 1.92 Å and 1.97 Å, respectively. So, Ga^{3+} ion has a smaller ionic radius compared to that of the Zn^{2+} ion. The length difference between Ga-O bond and Zn-O bond leads to stress of crystal and causes the stacking faults (Lee, 2013). In other words, the increase of Ga-doped concentration caused the stacking faults to be shorter and denser. Therefore, the Ga atoms in the ZnO crystal could induce stress between the Ga dopant and the original lattice host material.

Conclusion

We have successfully deposited ZnO:Ga thin films on corning glass by dc magnetron sputtering. The crystallinity of deposited films improved with the increase of Ga concentrations from 1 to 2%, then decrease at 3% Ga concentrations. The optical transmittance of films with 1% and 2% Ga concentration reach 85% in the visible range, while at 3% Ga concentration the transmittance of film only 70%. We observed that the band gap of films change due to the addition of Ga dopant. The band gap of films are 3.27, 3.28 and 3.21 eV for 1, 2 and 3% Ga-doped concentrations, respectively. Based on the structural and optical properties of the films, ZnO:Ga(2%) film is the best candidate for TCO application.

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Author's Contributions

Putut Marwoto: The leader of the study, data analysis and writing the manuscript.

Edy Wibowo: Data analysis and editing of the manuscript.

Dwi Suprayogi: Preparation and analysis of the sample.

Sulhadi Sulhadi: Preparation and analysis of the sample and contributed in the discussion part of the manuscript.

Didik Aryanto: Contributed in XRD analysis and the discussion part of the manuscript.

Sugianto Sugianto: Preparation of experiment apparatus, sample and data anaysis.

Ethics

This article is original. The corresponding author confirms that all of the other authors have read and approved the manuscript and no ethical issues involved.

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