



Available online at <http://journal.walisongo.ac.id/index.php/jnsmr>

Study of ZnO:Ga Thin Film Structure With Variation of Plasma Power Using the DC Magnetron Sputtering Method

Putut Marwoto¹, Budi Astuti¹, and Sugianto¹

¹Department of Physics, Faculty of Mathematics and Natural Sciences, Universitas Negeri Semarang, Indonesia

Abstracts

Corresponding Author:
pmarwoto@yahoo.com
Received: 17 September
2018, Revised : 20 Oktober
2018, Accepted: 27
November 2018

ZnO:Ga thin films grown on a corning glass substrate by the DC Magnetron Sputtering method have been successfully grown. The purpose of this study was to study the effect of plasma power on the resulting thin film structure. The quality and structure of the films were studied using X-ray diffraction (XRD), and scanning electron microscopy (SEM). Based on XRD characterization, it was found that the ZnO:Ga thin film has a hexagonal wurtzite structure on the C-axis with orientation planes (002) and (101). The quality of the resulting thin films can also be seen from the value of full-width at half maximum (FWHM) in the orientation plane (002) which increases with increasing plasma power during the growth process from 0.13°- 0.16°. The larger the FWHM value, the smaller the crystal grain size so that the structure becomes less good. This is supported by the surface morphology of the film which is less dense when the plasma power increases. ©2018 JNSMR UIN Walisongo. All rights reserved.

Keywords: ZnO:Ga thin film, structure, plasma power, DC magnetron sputtering

1. Introduction

ZnO materials are widely applied in electronic devices based on the unique properties of ZnO such as non-toxicity, good electrical properties, high transmittance and low manufacturing costs [1-4]. ZnO is a

material that has a direct band gap energy of 3.37 eV and a large exciton binding energy of about 60 meV at room temperature [5-6]. ZnO materials are widely applied as light emitting diodes, conductive electrodes, thin-film transistors, solar cells, sensors and so on [7-11].

The growth of ZnO material can use several techniques such as sol-gel, chemical vapor deposition, spray pyrolysis, RF sputtering and DC Magnetron sputtering [12-14]. Of these several techniques, DC Magnetron sputtering has advantages such as simple method, easy operation and affordable production price [15].

The properties of pure ZnO thin films are not good, such as high resistivity values and low transmittance [16]. This can be corrected by doping into the ZnO thin film. Materials from group IIIA such as aluminum, gallium and indium can be used as doping [17]. This is based on properties such as the atomic radius of Ga (0.062 nm) which is almost the same as the atomic radius of Zn (0.072 nm) so as to minimize the occurrence of defects in the resulting thin film [18]. Based on this, the quality of the ZnO:Ga thin films produced will be better, less reactive and resistant to oxidation.

In this article, it is reported about the growth of a ZnO:Ga thin film on a corning glass substrate using the DC Magnetron sputtering method. The effect of plasma power on the structure of the resulting film was studied using X-ray diffraction (XRD), and scanning electron microscopy (SEM).

2. Experiments Procedure

ZnO thin films were grown on corning glass substrate by DC Magnetron sputtering method. Preparation of ZnO:Ga targets. The targets were in the form of ZnO pellets with a purity of 99.999% and Ga₂O₃ with a purification of 99.999%. 99.99% argon (Ar) gas was used as sputter gas during the deposition process. Targets were made with a ratio of ZnO: 98 wt% and Ga₂O₃: 2 wt%. This research broadly includes four stages which can be seen in [19]. The growth parameters of ZnO:Ga thin films are shown in Table 1. deposited at a temperature of 300oC, plasma power of 30 Watt, argon gas pressure of 500 mTorr for 1 hour.

Table 1. Parameters of growth of ZnO:Ga thin films with variations in plasma power

| No. | Growth parameters | Condition |
|-----|------------------------|---------------------------|
| 1. | Base Pressure | 1 x 10 ⁻⁴ Torr |
| 2. | Growth Pressure | 500 mTorr |
| 3. | Deposition time | 120 menit |
| 4. | Plasma Power | 34,00 & 37,00 watt |
| 5. | Deposition Temperature | 300°C |

The phase of the ZnO:Ga thin film was determined by X-ray diffraction using a panalytical diffractometer, the wavelength used was 0.154 nm. The surface morphology and composition of the constituent elements of the resulting ZnO:Ga thin films were tested using scanning electron microscopy and energy dispersive x-ray spectroscopy with the phenom proX model.

3. Result and Discussion

The X-ray diffractogram for ZnO:Ga thin films grown on a corning glass substrate using the DC Magnetron sputtering method at varying plasma power is shown in Figure 1. Clearly, the thin films grown at 34 W plasma show two peaks. XRD at 2θ = 34.32° and 47.61° which are orientation planes (002) and (006), respectively [20]. Furthermore, when the plasma power was increased to 37 W, the XRD peak for the orientation plane (002) remained, but for the orientation plane (006) disappeared. It can be said that with the increase in plasma power in the crystal phase growth process, the ZnO:Ga thin film changes from polycrystalline to single crystal. However, this needs further research, considering that the method used is sputtering which is a type of physical growth method and it is very difficult to get crystals with single crystal properties. The XRD peaks produced on ZnO:Ga thin films on corning glass substrates are oriented in the C-axis with a hexagonal wurtzite phase [21].

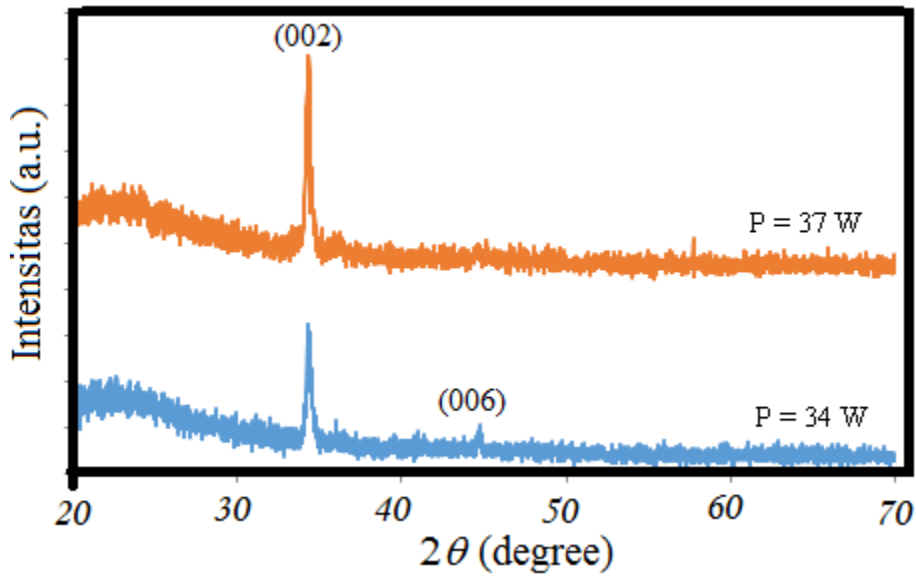


Figure 1. X-ray diffraction form of a ZnO:Ga thin film sample with variations in plasma power

Table 2. X-ray diffraction analysis results for ZnO:Ga thin films at variations in plasma power

| Sample | Plasma Power (W) | Angel 2θ (°) | Peak FWHM (002) (°) | <i>d</i> -space (Å) | Grain crystal size (nm) |
|--------|------------------|---------------------|---------------------|---------------------|-------------------------|
| #A | 34 | 34,32 | 0,1338 | 2,6129 | 9,162 |
| #B | 37 | 34,30 | 0,1632 | 2,6120 | 6,926 |

Based on Figure 1, it can be determined the crystal grain size for the XRD peak (002) by using the Debye-Scherer formula [22]:

$$d = \frac{c\lambda}{\beta \cos\theta} \tag{1}$$

Where *d* is the crystal grain size, is the full width at half maximum (FWHM) of the XRD peak (002), is the X-ray wavelength (= 0.154 nm), is the Bragg angle and *c* is the correction factor for thin films that magnitude is 0.94 . The results of the XRD analysis also showed that the diffraction angle, FWHM, *d*-spacing, and crystal grain size are shown in Table 2.

Based on Table 2, it can be seen that with the addition of plasma power on the growth of ZnO:Ga thin films, the crystal quality decreases. This is indicated by the smaller crystal grain size. It is known that a good crystal is a crystal that has a narrow FWHM value so that the crystal size becomes

large, as a result the grain boundary will become larger [23]. The grain boundary is the surface or area that connects two single crystal grains. This is contrary to [24], which states that increasing plasma power will decrease the FWHM value so that the crystal grain size will increase so that the crystal quality will be better. Figure 1 also shows that the 2θ angle does not shift, which means that the Ga doping on the ZnO thin film does not change the X-ray diffraction and does not give rise to new diffraction peaks which are the diffraction peaks of element Ga.

Furthermore, using the crystal grain size data above, it is possible to calculate the dislocation density (δ) which is defined as the distance or length of the dislocation line per unit volume. The dislocation density (δ) is calculated using equation [25]:

$$\delta = \frac{l}{d^2} \tag{2}$$

And the strain (ϵ) of the ZnO:Ga thin film can be calculated using the equation:

$$\epsilon = \frac{\beta \cos \theta}{4} \quad (3)$$

The results of structural parameter analysis of ZnO:Ga thin films for dislocation density and strain are shown in Table 3.

Table 3. Structural parameters such as dislocation density, δ and strain, ϵ

| Sample | Plasma Power (W) | $\delta(\times 10^{15})$ (line/m ²) | $\epsilon (\times 10^{-3})$ |
|--------|------------------|---|-----------------------------|
| #A | 34 | 11,90 | 3,95 |
| #B | 37 | 20,80 | 5,23 |

It can be seen from Table 3, the strain value increases with increasing plasma power. Increasing the value of strain will cause the dislocations to move towards the surface of the film so that deformation will occur. During movement, these dislocations will interact with each other. The result of the interaction is that there is one that makes movement easier but some makes it difficult to move. The result of the reaction between dislocations which complicates the movement, actually serves as a source of new dislocations so that the dislocation speed increases. As a result, the dislocation density increases so that the dislocation movement will be difficult to move. Finally, the difficulty of moving due to the large dislocation density will increase the strength of the film [26].

The surface morphology of the ZnO:Ga thin films for varying plasma power is shown in Figure 2. It is clearly seen in Figure 2, that the crystal grain size decreases when the plasma power is increased. This supports the results of the XRD analysis. The decrease in crystal grain size with increasing plasma power can be said that the quality of the resulting film decreases []. Based on this, it can be said that the plasma power of 34 W

has better crystal quality than the film grown at the plasma power of 37 W.

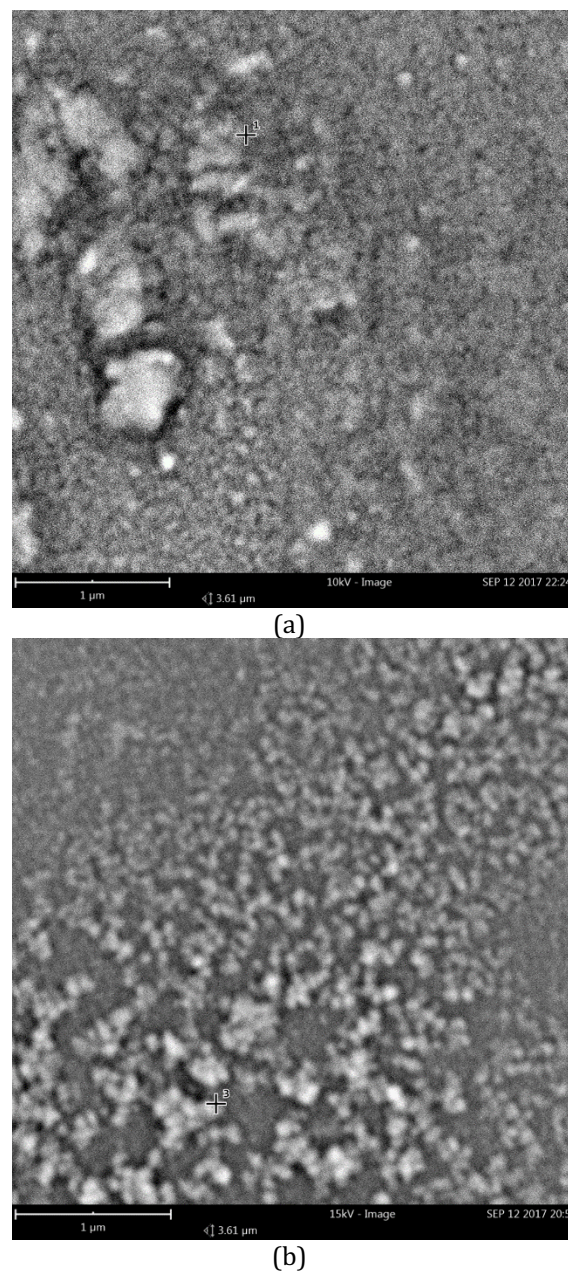


Figure 2. Surface morphology of ZnO: Ga thin films at variations in plasma power (a) 34 W and (b) 37 W

4. Conclusion

The growth of ZnO:Ga thin films on corning glass substrates using the DC Magnetron sputtering method has been successfully carried out. The resulting ZnO:Ga thin films were studied using XRD to study the crystal structure and to study the surface morphology by SEM. The resulting

thin films showed a decrease in crystal quality with increasing plasma power in the growth process. This is indicated by a decrease in the crystal grain size which causes an increase in the dislocation density and strain of the resulting film.

Acknowledgment

This research was supported by the DIPA program at the Faculty of Mathematics and Natural Sciences, Semarang State University from the finance of the Kemenristek Dikti project in 2017. The authors also thank Agus Andi Wibowo, S.Si who has helped technical growth, Natalia Erna, S.d. and Rodhotul Mutaqin, S.Si. who helped during the characterization process.

References

- [1] M. Osada, T. Sakemi, T. Yamamoto, The effect of oxygen partial pressure on local structural properties for Ga-doped ZnO thin films, *Thin Solid Films*, 494, 38-41, 2006.
- [2] C. Huang, D. Chen, C. Hsu, Influence of deposition parameter and annealing treatment on the properties of GZO films grown using rf magnetron sputtering, *Ceramics International*, 38, 1057-1063, 2012.
- [3] S. Kim, W.I. Lee, E.-H. Lee, S. Hwang, C. Lee, Dependence of the resistivity and the transmittance of sputter-deposited Ga-doped ZnO films on oxygen partial pressure and sputtering temperature, *Journal of materials science*, 42, 4845-4849, 2007.
- [4] N. Habubi, S. Chiad, S. Jabbar, W. Jabbar, Synthesis and optical properties of sprayed ZnO and ZnO:Ga thin films, *Journal of the Arkansas Academy of Science*, 66, 82, 2012.
- [5] Balta, A.K. Ertek, O. Eker, N. & Okur, I. 2015. MgO and ZnO Composite Thin Films Using Thin Spin Coating Method on Microscope Glasses. *Material Sciences and Applications*. 6:40-47.
- [6] Shariffudin, S.S. Mamat, M.H. & Rusop, M. 2010. Post Annealing Temperature Effect On Photoluminescence Spectroscopy of ZnO Thin Film. *2010 International Conference on Electronic Devices System and Application (ICEDSA 2010)*. IEEE: 360-363.
- [7] Ebrahimifard, R. Golobostanfard, M.R. Abdizadeh, H. 2014. Sol Gel Derived Al and Ga c-doped ZnO Thin Film: An Optoelectronics Study. *Applied Surface Science*. 290:252-259.
- [8] M. Wu, S. Yu, L. He, G. Zhang, D. Ling, W. Zhang, Influence of oxygen pressure the structural, electrical and optical properties of Nb-doped ZnO thin films prepared by pulsed laser deposition, *Applied Surface Science*, 292, 219-224, 2014.
- [9] S. Kim, W.I. Lee, E.-H. Lee, S. Hwang, C. Lee, Dependence of the resistivity and the transmittance of sputter-deposited Ga-doped ZnO films on oxygen partial pressure and sputtering temperature, *Journal of materials science*, 42, 4845-4849, 2007.
- [10] H.H. Shin, Y.H. Joung, S.J. Kang, Influence of the substrate temperature on the optical and electrical properties of Ga-doped ZnO thin films fabricated by pulsed laser deposition, *Journal of Materials Science: Materials in Electronics*, 20, 704-708, 2009.
- [11] Y. Xue, H. He, Y. Jin, B. Lu, H. Cao, J. Jiang, S. Bai, Z. Ye, Effect of oxygen plasma treatment on the surface properties of Ga-doped ZnO thin films, *Applied Physics A*, 114, 509-513, 2013.
- [12] T.P. Rao, M.S. Kumar, N.S. Hussain, Effect of thickness and atmospheric annealing on structural, electrical and optical properties of GZO thin films by spray pyrolysis, *Journal of Alloys and Compounds*, 541, 495-504, 2012.
- [13] D.-S. Kim, J.-H. Park, S.-J. Lee, K.-J. Ahn, M.-S. Lee, M.-H. Ham, W. Lee, J.-M. Myoung, Effect of oxygen concentration on the properties of Al-doped ZnO transparent conductive films deposited by pulsed DC magnetron sputtering, *Materials Science in Semiconductor Processing*, 16, 997-1001, 2013.
- [14] Lupan, O., T. Pauporte, L. Chow, B. Viana, F. Pelle, L.K. Ono, B.R. Cuenya, & H. Heinrich. 2010. Effects of annealing on properties of ZnO thin films prepared by electrochemical deposition in chloride

- medium. *Applied Surface Science*, Vol. 256, pp.1895–1907.
- [15] Ismail, A. & Abdullah, M.J. 2013. The structural and optical properties of ZnO thin films prepared at different RF sputtering power. *Journal of King Saud University – Science*, Vol. 25, pp.209–215.
- [16] Li, Ying, Huang Qin, Xiaofang Bi. 2013. Stress Dependent Properties of Ga-Doped ZnO Thin Films Prepared by Magnetron Sputtering. *Journal Mater Sci*, 24. 79-84.
- [17] Suhea, M., S. Christoulakis, N. Katsarakis, T. Kitsopoulos, & G. Kiriakidis. 2007. Comparative Study of Zinc Oxide and Aluminum Doped Zinc Oxide Transparent Thin Films Grown by Direct Current Magnetron Sputtering. Elsevier. *Thin Solid Films*, 515, 6562-6566.
- [18] J.-H. Yang, H.-S. Kim, J.-H. Lim, D.-K. Hwang, J.-Y. Oh, S.-J. Park, The effect of Ar/O₂ sputtering gas on the phosphorus-doped p-Type ZnO thin films, *Journal of The Electrochemical Society*, 153, G242-G244, 2006.
- [19] Sugianto, Zannah, R. Mahmudah, S.N. Astuti, B. Putra, N.M.D. Wibowo, A.A. Marwoto, P. Aryanto, D. & Wibowo, E. 2016. Pengaruh Temperatur Annealing Pada Sifat Listrik Film Tipis Zinc Oksida Doping Aluminium Oksida. *Jurnal MIPA*. 39(2): 115-122.
- [20] C. periasamy, R. Prakash, & p. Chakrabarti, 2009. Effect of post annealing on structural and optical properties of ZnO thin film deposited by vacuum coating technique. *J. Matel. Sci.: Mater Electron*. Springer.
- [21] Z. Shirui, D. Yabin, Yu Mingyan, G. Xiaolong, Xu Xinwei, J. Yupeng, & X. Yang. 2014. Microwave annealing effect on ZnO films deposited by atomic layer deposition. *J. Of Semicondutor*. 35(11):1-4.
- [22] Z.R. Khan, M. Zulfequar, & M.S. Khan. 2010. Optical and structural properties of thermally evaporated cadmium sulphide thin films on silicon (100) wafer. *Materials Science and Engineering*. B. 274: 2-3.
- [23] Suryanarayana C & M. Grant Norton. 1998. *X-ray Diffraction A Partical Approach*. New York: plenum Press.
- [24] B.S. Li, Y.C. Liu, D.Z. Shen, J.Y. Zhang, Y.M. Lu. & X.W. Fan. 2003. Effects of RF power on properties of ZnO thinfilms grown on Si(100) substrate by plasma enhanced chemical vapor deposition. *J. Cryst. Growth*. 249:179-185.
- [25] Z.R. Khan, M.S. Khan, M. Zulfequar, & M. S. Khan. 2011. Optical and structural properties of ZnO thin films fabricated by sol-gel method. *Mat. Sciences and Appl*. 2:340-345.
- [26] P. Schall, M. Feuerbacher, M. Bartsch, U. Messerschmidt, & K. Urban. 1999. Dislocation density evolution upon plastic deformation of Al-d-Mn single quasicrystals. *Hilospical Magaine Letters*. 79(10):785-796.