Effects of Argon Pressure on the Properties of ZnO:Ga Thin Films Deposited by DC Magnetron Sputtering

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Effects of Argon Pressure on the Properties of ZnO:Ga Thin Films Deposited by DC Magnetron Sputtering

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Abstract. Gallium (Ga)-doped zinc oxide (ZnO:Ga) thin films were deposited on corning glass substrates by homemade DC magnetron sputtering. Effects of argon gas pressure on the structural and optical properties of ZnO:Ga thin films were investigated by XRD, SEM and UV-Vis spectroscopy. The argon gas pressure was adjusted at 450, 500 and 550 mtorr. All the films exhibit a strong (002) peak and a weak (004) peaks. The XRD pattern demonstrated that crystallinity of the film improved with increasing of the argon pressure. ZnO:Ga thin films deposited have crystalline structure. It was shown that the argon pressure has a great influence on ZnO:Ga film surface structures. The grain size of the films was increased with the increases of argon pressure. The grains shape of the film change from an equiaxed rough grain to a longish grain with the argon pressure. The average of transmittance of the films is about 80% in the visible range. It is shown that the argon pressure has no effect significantly on optical bandgap of ZnO:Ga, but in general it can be explained that increasing of the argon pressure can reduce the bandgap. The optical bandgap of ZnO:Ga thin films in the range of 3.25 - 3.3 eV.

Keywords: ZnO:Ga, DC magnetron sputtering
PACS: 81

INTRODUCTION

Zinc oxide (ZnO) is a promising materials for application in electronics and optoelectronics devices such as transparent electrodes, sensors, window materials for display and solar cells. Therefore, ZnO has attracted great attention fo few years. Furthermore, ZnO is also a materials alternative in transparent conductive oxides (TCO) applications [1,2], due to its low cost and relatively low deposition temperature [1]. However, the properties of the pure ZnO are unstable [3] and highly resistive [4]. To improve these properties, ZnO needs to be doped with other materials, especially on the device application purposes[4]. Doping is one of the powerful methods to control electrical properties [5]. ZnO was frequently doped with group III elements such as critical properties B, In, Al and Ga [2]. Ga is reactive and also the most effective n-type dopant for ZnO because the rather similar radius compared with zinc (Zn) [5]. So, the covalent bond of Ga-O and Zn-O are nearly equal [3].

Galium doped zinc oxide (ZnO:Ga) films have been deposited by using several techniques, such us reactive magnetron sputtering [1,2,6], physical vapor deposition [5], sol-gel [7], atomic layer deposition [8] and spray pyrolysis [4] and the other method. Among the deposition techniques, DC magnetron sputtering method is promising in preparing ZnO:Ga thin films due to the low cost of the source materials preparation, and high growth rates [1]. Moreover, sputtering methods have several advantages such as large area films and the growth rates can be easily-controlled during growth process [6].

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In the previous work, we has been done on the growth of ZnO:Ga thin films using home made DC magnetron sputtering [9]. Furthermore, in this work, we reported the effect of the argon pressure on the ztructural and optical properties of ZnO:Ga thin films by using DC magnetron sputtering.

EXPERIMENTAL PROCEDURE

ZnO:Ga thin films were deposited on corning glass substrates by *homemade* DC magnetron sputtering technique [9,10]. Ga_2O_3 (99.999%) and ZnO (99.999%) powder was mixed and pressed as a source target with total mass of 10 gram. The Ga doping was fixed at 2 % in present work. The diamater of the target was 2.5 cm. Corning glass substrates were cleaned with aceton and methanol solution using ultrasonic bath for 15 minutes. The substrate temperature, sputtering power, and deposition time were kept constant at 400°C, 30 W and 60 minutes, respectively. The argon gas pressure was varied at 450, 500 and 550 mtorr.

The crystal structure properties and surface morphologies of the films were analyzed by X-ray diffraction with $Cu-K_\alpha$ radiation (1.5406 Å) and scanning electron microscopy (SEM), respectively. The optical properties was measured by ultraviolet spectrocopy (UV-Vis).

RESULT AND DISCUSSION

XRD pattern of ZnO:Ga films deposited on corning glass substrate with different argon pressure is shown in Fig. 1. All the films exhibit a strong (002) peak and a weak (004) peaks. However, the films deposited at 400 and 550 mtorr argon pressure have a weak (101) peak. The ZnO:Ga deposited films are polycrystalline with peak oriented along the (002) plane. It indicated that the grains of the films were strongly oriented along the *c*-axis of the wurtzite structure [5]. The XRD pattern demonstrated that crystallinity of the film was improved with increasing of the argon pressure. In other words, the wurtzite-type crystal structure of ZnO could be formed to produce the ZnO:Ga films at any argon pressures. The (002) peak of the film prepared at 550 mtorr argon pressure is the highest among the samples. It revealed that the ZnO:Ga film deposited at 550 mtorr has the highest crystallinity of any film produced at argon pressure between 450 – 550 mtorr.

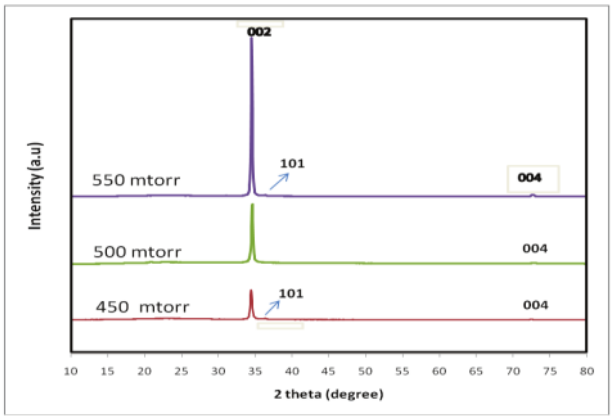


FIGURE 1. X-ray diffraction patterns of ZnO:Ga films deposited at different argon pressures.

The XRD patterns also shows, no diffraction Ga_2O_3 phase were detected. Nevertheles, it is believed that Ga atoms substitute Zn in hexagonal lattice and form Ga-O bond. It can be explained that Ga atoms are ionized into Ga^{3+} and subsitute Zn^{2+} [1], which makes the *c*-parameter shorter due to the fact that ionic radius of Ga is smaller than that of Zn [2]. The addition of Ga concentration also was studied by other researcher such as, Lee et al. [5] was reported that the increasing of Ga concentration showed a broad peak (002). Recently, Maeng and Park [8] also reported that the peak intensity is significantly decreased with increasing Ga concentration.

TABLE 1. Structure parameters of ZnO:Ga thin films deposited at different argon pressures

Argon pressure (mtorr)	2θ (°)	Lattice constant c (Å)	Crystal size (nm)	d-spacing (Å)
450	34.44	5.20	*43*	*2.60*
500	34.44	5.20	*24*	*2.60*
550	34.50	5.19	*32*	*2.59*

From Fig. 1, it is demonstrated that the intensity of the (002) peak was enhanced and becomes more intense and sharper as the argon gas pressure increases. The intensity (002) peak at 450 mtorr argon pressure is very weak, which can be explained to the degrade crystallinity. It was also observed that the Bragg angle 2θ of ZnO:Ga (002) at 450, 500 and 550 mtorr argon pressure is 34.44, 34.44 and 34.50, respectively. This result shown that the position of the (002) peak at 550 mtorr argon pressure was shifted towards higher 2θ values. The sifted implied that the lattice constant decreased [6].

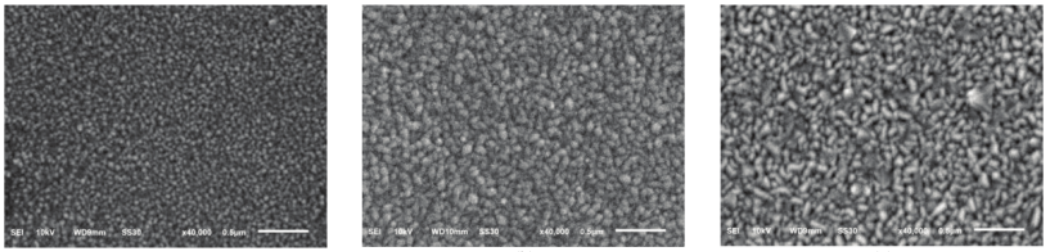
Table 1 shows the calculated structure parameters of ZnO:Ga films deposited. The crystal size was obtained by using the Scherer formula

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

where D is the diameter of the crytallites film, λ is the wavelength of $Cu-K_\alpha$ line (1.5406 Å), β is FWHM and θ is the Bragg angle. The standard ZnO has the Bragg angle 2θ at 34.45°, with lattice constant c at 5.21 Å [2]. Therefore, the ZnO:Ga thin films deposited at 550 mtorr argon pressures present a larger the Bragg angle 2θ (34.50°) and smaller lattice constants (~5.19 Å). It can be revealed that 550 mtorr argon pressure makes the lattice constant c of the ZnO:Ga thin film shorter. This case is similar with Ga (1%) doped ZnO as reported by Fang et al.[2].

The effect of argon pressure on the ZnO:Ga thin film morphology was observed by using SEM micrographs. Figure 2 shows the surface morphology of films at three different argon pressures: (a) 450, (b) 500, and 550 mtorr, respectively. Its clearly shown that the argon pressure has a great influence on ZnO:Ga film surface structures, which surface roughness increases with the argon pressure was increased. Ma et al. [1] reported that surface roughness of the ZnO:Ga strongly depend on the deposition pressure.

From Fig. 2(a), the ZnO:Ga thin film deposited at 450 mtorr shows the smooth surface. It is found that the grains are entirely dense and stacked up uniformly with diameter about 50 nm. In fact, a low argon pressure causes



(a) (b) (c)

FIGURE 2. SEM morphologies of ZnO:Ga thin films deposited with different pressures: (a) 450 mtorr, (b) 500 mtorr, and (c) 550 mtorr

the amount of Ar^+ ions are also low, so the particles from the sputerring target had large energy to collide with the substrate. This condition was caused partially of the particles would be rebounced, and then the surface roughness decreased. Furthermore, at high energy, the collisions between Ar^+ ion and the sputtered particles caused the oxygen atoms may be released from the Zn-O or Ga-O bonding, so that the crystal quality degraded, as shown in Fig. 1. Figure 2(b) shows surface morphology of the film grown at 500 mtorr with the grain diamters of 100 nm. At higher argon pressure, the particles from sputtered target enhanced as increasing Ar^+ ions. Therefore, the sputtered particles had low energy when it gets to the substrate surface. This condition was caused the grains of the sample

was slowly grown, so the surface of the film becomes more roughness. The grain size tends to increase and the grain shape tends to change from an equiaxed rough grain to a longish grain [11] when the argon pressure increases from 500 to 550 mtorr. It is shown in Fig. 2(c) that the ZnO:Ga thin film deposited at 550 mtorr film has the elongation grain to about 150 nm.

Figure 3 shows the transmittance of the ZnO:Ga thin films prepared at different argon pressures as a function of the wavelength in the visible region. The average optical transmittance of all films is around 80% in the visible region. The transmittance curve in Fig. 3 shows steeper graph when the argon pressure was increased. This can be attributed that the structural homogeneity and crystallinity of the films was improved with the argon pressure increases [4]. This result consistent with XRD pattern as presented in Fig. 1. Furthermore, it is found that the film deposited at 550 mtorr argon pressure had an improved transmittance of about 85% in the range of 400 – 800 nm (visible region).

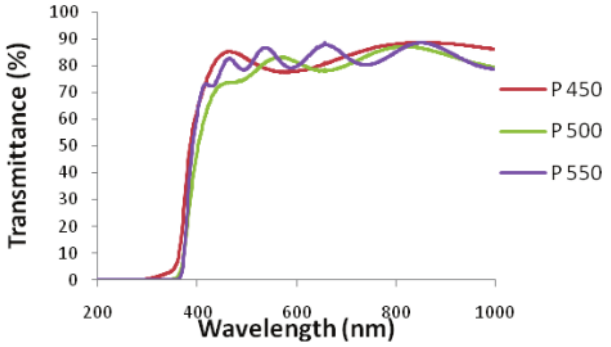


FIGURE 3. Transmittance of ZnO:Ga thin films prepared at different argon pressures.

The optical energy band gap can be determined from the plotting α^2 vs. $h\nu$ where α is the absorption coefficient and $h\nu$ is the photon energy, such kind of plots are shown in Fig. 4. The calculated valued of optical band gap (E_g) of ZnO:Ga films deposited at 450, 500 and 550 mtorr are presented in Table 2. Maeng and Park [8] reported that the band gap width can affect the transmittance. The minimum optical bandgap (3.25 eV) could be found at 500 mtorr argon pressure. It is shown that the argon pressure has no effect significantly on optical bandgap of ZnO:Ga, but in general it can be explained that inreasing the argon pressure can reduce the bandgap. Ma et al. [1] reported that reducing bandgap is due to carrier concentration effect. The decreasing of energy bandgap is known as "red-shift" [2]. In addition, the broadening of the energy bandgap which is attributed to increase of carrier concentration as known as Burstein-Moss shift (B-M effect) [8].

TABLE 2. Energy bandgap of ZnO:Ga thin films deposited at different argon pressures

Argon pressure (mtorr)	Energy bandgap (eV)
450	3.33
500	3.25
550	3.30

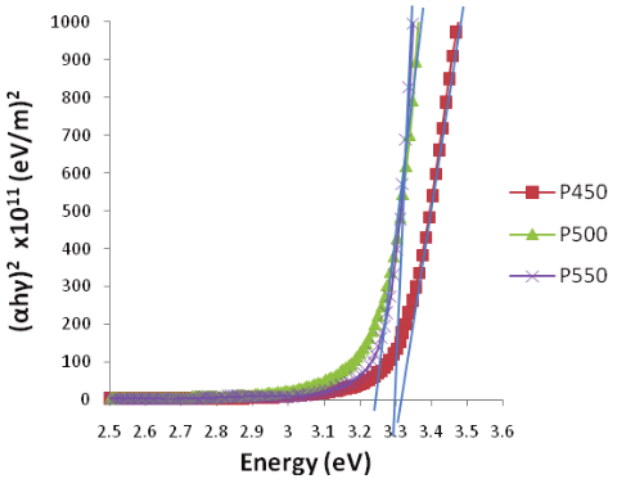


FIGURE 4. Plot of $(\alpha h\nu)^2$ as a function of photon energy for ZnO:Ga thin films prepared at different argon pressures.

CONCLUSIONS

ZnO:Ga thin films were deposited on corning glass substrates by homemade DC magnetron sputtering. The films were grown at 450, 500 and 550 mtorr argon pressures. Effects of argon gas pressure on the structural and optical properties of ZnO:Ga thin films were investigated by XRD, SEM and UV-Vis spectroscopy. The XRD pattern demonstrated that crystallinity of the film improved with increasing of the argon pressure. The ZnO:Ga films are polycrystalline. The argon pressure has a great influence on ZnO:Ga film surface structures. The grain size of the films tend to increase and the grain shape tends to change from an equiaxed rough grain to a longish grain when the argon pressure increases. The transmittance curve has steeper graph with an increase transmittance when argon pressure was increased. The average of transmittance of the films is about 80% in the visible range. Althought, experiment shown that the argon pressure has no effect significantly on optical bandgap of ZnO:Ga, but in general it can be explained that inreasing the argon pressure can reduce the bandgap. The optical bandgap of ZnO:Ga thin films is about 3.25 - 3.3 eV.

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