Investigation of growth of ZnO thin films via RF sputtering system and in-situ post annealing

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Abstract

The present article deals with in situ post annealing of ZnO in sputtering system. The ZnO thin films were grown at low temperature of 100℃ and at working pressure of 15 mTorr with RF magnetron sputtering. Having been gown, ZnO thin films were annealed in situ at different temperatures, at annealing ambient pressure of 15 mTorr and in ambients of oxygen and argon respectively . Through analyses of XRDs, it is can be concluded that the crystallinity of annealed ZnO thin films becomes much better than that of as-grown ZnO thin film.

Key Words : sputtering system, in-situ post annealing, annealing ambient pressure

1. Introduction

Zinc oxide is a wide band gap material with a high chemical stability, good photoelectric and piezoelectric properties[1]. Single crystal ZnO films with good quality are imperative to devices of surface acoustic wave and photoelectronics. There are various fabrication methods of ZnO thin films[2-4]. In order to get ZnO thin film with high quality, it is comprehensive to use post annealing methods, such as rapid thermal annealing(RTA)[5], vacuum annealing and various furnace annealing. We attempted to use in situ post annealing method to get high quality ZnO thin films.

2. Experimental process

We grew ZnO thin films by using RF magnetron sputtering and chose (100)-oriented silicon wafers as substrates, exploited 5N ZnO for target and set substrate temperature at 100℃. The substrates were ultrasonically cleaned in acetone and ethanol during 10 min, rinsed in deionized water, and subsequently dried with nitrogen gas before being introduced in the sputtering system. The growth working pressure was 15 mTorr and ambient was mixture of Ar (5N purity) and O_2(5N purity) whose ratio was 3:2. After growth the sputtering chamber was evacuated to below 10^{-5} Torr and then was introduced with Ar or O_2 to the annealing ambient pressure of 15 mTorr again, setting substrate temperatures at 600℃ and 800℃ respectively. We took 180 minutes for thin film growth and 30 minutes or different other minutes for post annealing times. Then we conducted XRDs and DFM on samples to study the crystallinities of ZnO.

3. Results and discussion

Fig.1 is XRDs of as-grown ZnO thin film(a) and annealed ZnO thin films at 600℃(b, e) and 800℃(c, e) at all of which 30 minutes was holden. Fig. 1 shows that except on the condition of annealing temperature of 800℃ in air furnace, (0002) peaks of ZnO can be seen and this means that nearly all ZnO thin films have c-axis-oriented growth. Fig.1 also shows that the sample annealed in air furnace for only 30 minutes holding time has not any peaks. That means the sample got amorphous. Perhaps the ZnO thin film was ablated by bombardments of hot ambient molecules with high thermal energy. Intensities of (0002) peaks of others show that as-grown film has worst crystallinity and annealing treatments promote crystallinity except at too high temperature in furnace, especially at 800℃ in Ar.

Table 1 shows that the values of (0002) peak positions of annealed ZnO thin films have shifts bigger than that of as-grown ZnO thin film. The higher annealing temperature is, the more shift is. And also the effect of shift in Ar is more than that in O_2 ambient. FWHMs on table 1 show that high post annealing temperatures makes values of FWHMs smaller. It implies from above results that post annealing makes the stress of films relaxed, makes defects, such as O vacancy and zinc interstitial, destroyed and makes crystal grains of films bigger. But relaxation in 30

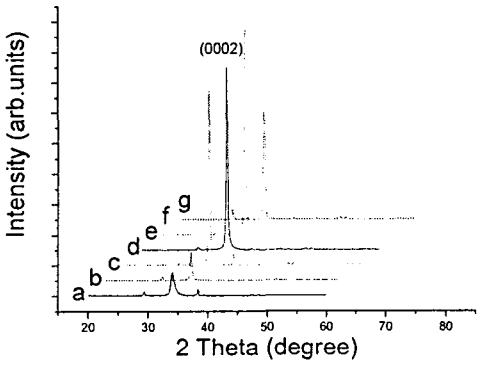

Fig.1. The XRDs of ZnO thin films grown on the condition of (a) as-grown and annealed at substrate temperatures of (b) 600℃ in O_2, (c) 800℃ in O_2, (d) 600℃ in Ar, (e) 800℃ in Ar, (f) 600℃ in furnace and (g) 800℃ in furnace.

Table 1. The (0002)-peak positions and FWHMs of XRDs of ZnO films

condition	(0002) position	FWHM[°]
as-grown	34.06	0.68
600℃ in O_2	34.18	0.38
800℃ in O_2	34.22	0.30
600℃ in Ar	34.20	0.30
800℃ in Ar	34.23	0.30
600℃ in fur.	--	0.44

minutes holding time isn't sufficient.

We prepared ZnO thin films as above and annealed in-situ at temperature of 800°C at Ar ambient pressure of 15 mTorr in the annealing periods of 80min., 120min. and 180min. respectively. All samples annealed in different annealing holding periods showed (0002) peaks from XRDs.

Table 2 shows that the prolonged annealing holding time made (0002) peak shift to be bigger glanced angle and made FWHMs turn smaller. But for too prolonged annealing holding time of 180 minutes, the (0002) peak comes back smaller a little and FWHM bigger. From Table 2 it is implied that prolonged annealing holding time makes ZnO films relaxed more sufficiently. But too prolonged time makes ZnO thin films to be damaged a little because of unobvious reasons.

Dynamic forced microscopes(DFMs) of samples show that high temperature make surfaces of samples smoother. The higher the temperature is, the smoother the surface is. From the photos in Fig.2, we can't tell the difference of roughness between surface of annealed at 800°C in Ar and in oxygen, though from the (0002) peak positions on Table 1, the thin film annealed at 800°C in Ar has a little more relaxation than that annealed at 800°C in oxygen.

4. Conclusion

The ZnO thin films grown at low temperature by RF magnetron sputtering and annealed in-situ at high temperature at low annealing ambient pressure have higher crystallinity than that at same temperature in air furnace. Within short annealing period, the higher the annealing temperature is, the better the crystallinity of ZnO thin films is. Inert ambient gas is more effective than active gases in-situ post annealing at same temperature. In-situ post annealing makes c-axis-grown ZnO grain bigger and also makes surface of thin films smoother. In order to get good quality ZnO thin films at high annealing temperature, appropriate time period is needed. The method is convenient to get high quality ZnO thin films.

Table 2. *Effects of different annealed times*

condition	(0002) position	FWHM[°]
80 min. in Ar	34.21	0.29
120min. in Ar	34.35	0.27
180min. in Ar	34.32	0.34

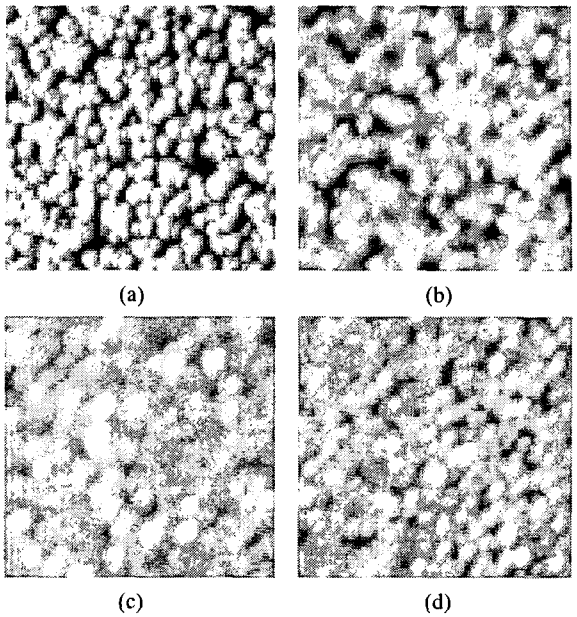

Fig.2. The DFMs of samples of (a) as-grown, (b) annealed at 600℃ in Furnace, (c) 800℃ in O_2 ambient and (d) 800℃ in Ar ambient

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6. References

[1] T. K. Subramanyam, B. Srinivasulu Naidu, S. Uthanna, Crystal Research and Technology, Vol. 8, No. 34, p. 981, 1999.

[2] S. Hayamizu, H. Tabata, T. Hawai, J. Appl. Phys. Vol. 80, p. 787, 1996.

[3] K. Iwata, P. Fons, S. Niki, A. Yamada, K. Nakahara, T. Tanabe, H. Takasu, J. Cryst. Growth, Vol. 214-215, p. 50, 2000.

[4] J. W. Lim, H. S. Park, S. W. Kang, J. Appl. Phys. Vol. 88, p. 6327, 2000.

[5] S. T. Shishiyanul, O. L. Lupan, E. V. Monaico, V. V. Ursaki, Thin Solid Films, Vol. 488, p. 15, 2005.