Surface Morphology and Electrical Properties of ZnO:Ga Films Formed by Magnetron Sputtering

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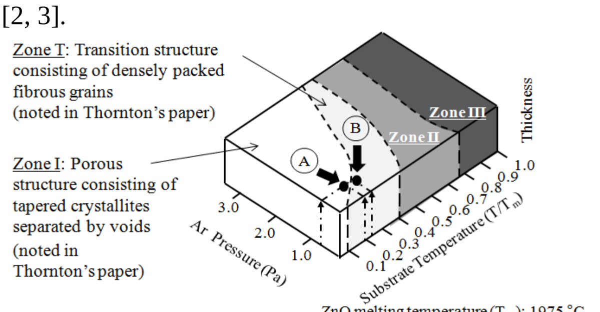
1. Introduction

Optically transparent and conductive ZnO films, such as Ga-doped Zinc oxide (ZnO:Ga) or Al-doped ZnO (ZnO:Al) films, are attracting attention as alternative electrode materials to indium tin oxide (ITO) films.

We have reported that Thornton's zone model [1], as shown in Fig. 1, can be applied to the relationship between the ZnO:Ga film microstructure and the dc magnetron sputtering conditions [2, 3]. In the report, the resistivity of films thinner than ca. 200 nm formed at 250 °C (Ⓑ: the point in Zone T close to the boundary with Zone I in the map of Thornton's model) was found to decrease with film thickness. In contrast, the resistivity of films thicker than ca. 200 nm increased with the thickness, as shown in Fig. 2(a). In this work, the cause of the increase in resistivity with thickness in region-2 (Fig. 2) was investigated. In addition, the resistivities of the films formed at 140 °C (Zone I, Fig. 1) decreased monotonically with film thickness, even for films thicker than ca. 200 nm. The films in Zone I had a pebble-like surface morphology, as shown in the micrograph of Fig. 3(a), and the pebble size increased with the film thickness [2, 3].



ZnO melting temperature (T_m): 1975 °C

Ⓐ Substrate Temperature: 140 °C, Pressure: 0.666 Pa, $T/T_m=0.18$

Ⓑ Substrate Temperature: 250 °C, Pressure: 0.666 Pa, $T/T_m=0.23$

Fig. 1. Schematic diagram of the relationship between the microstructure of a film and the sputtering conditions..

2. Experimental

ZnO:Ga films were formed using a conventional magnetron sputtering system under dc power (USP-66F, Universal Systems Co.). The electrical properties of the ZnO:Ga films were analyzed from Hall effect measurements conducted using the van der Pauw technique (HL5500PC, Accent Co.). The film nanostructure was observed using a scanning electron microscope (SEM; SU8020, Hitachi High-Technologies Co.) and a transmission electron microscope (TEM; H-9000UHR, Hitachi High-Technologies Co.).

3. Results and Discussion

The film produced with a substrate temperature of 250 °C under an operating (sputtering) Ar pressure of 0.666 Pa corresponds to the point near the border line in Zone T of Thornton's Zone model (Fig. 1). The resistivities of the films were not entirely dependent on the Ga_2O_3 concentration of the sputtering target in the range of 4.0-6.0 wt%, as shown in Fig. 2(a). The films thicker than ca. 200 nm (region-2 in Fig. 2) had different morphologies from those less than ca 200 nm (region-1) as shown by the SEM micrographs in the figure. The surface morphologies of the films in region-2 appear porous, whereas the films in region-1 have morphologies consisting of densely stacked grains. A cross-sectional TEM of a 925 nm thick film provides evidence that the microstructure of the films in region-2 are changed significantly from the densely packed structures in region-1 to a porous form, as shown in Fig. 2(b).

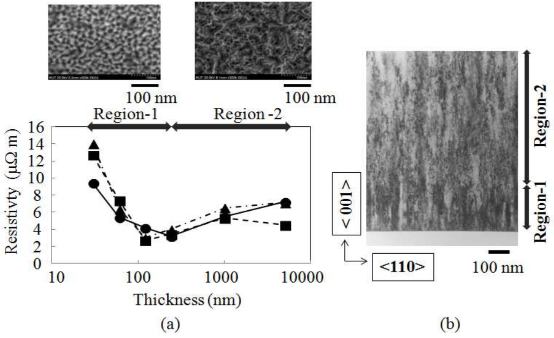


Fig. 2. (a) Resistivity of ZnO:Ga films as a function of thickness formed using sputtering targets with various Ga_2O_3 concentrations: ■ 4.0 wt%, ▲ 5.0 wt%, and ● 6.0 wt%. SEM micrograph shows the typical surface morphology of the film in each region. (b) TEM cross sectional micrograph of the ZnO:Ga film prepared using a 6.0 wt% Ga_2O_3 target.

This change in microstructure was also analyzed with respect to the resistivity that resulted from different substrate temperatures used during the deposition process, as shown in Fig. 3. The film formed at 250 °C (Fig. 3(b)) has a morphology consisting of densely stacked grains. In contrast, the film deposited at a substrate temperature of 290 °C (Fig. 3(c)) had a porous morphology. The growth of such porous structure was attributable to the development of surface agglomeration caused by the surface tension or the surface fluidity of the film during the deposition process at the substrate temperature. From the comparison among the micrographs shown in figure 2 and in figure 3, it was concluded that the porous film structure formed in region-2 (Fig. 2) was caused by surface agglomeration according to heating of the substrate from 250 °C to ca. 290 °C by irradiation of plasma during the deposition process. In addition, the surface morphologies of the non-doped ZnO films were not dependent on the substrate temperature at less than 300 °C and no agglomeration was observed. Then, the surface agglomeration phenomenon of the ZnO:Ga film at ca. 300 °C substrate temperature is closely related to the doping of Ga into ZnO.

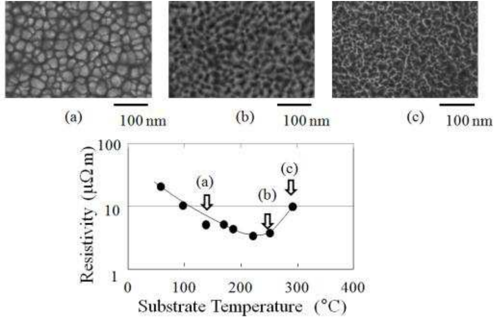


Fig.3. Resistivity and surface morphology of ZnO:Ga films as a function of substrate temperature. Films with thicknesses from121 to 132 nm were formed using ZnO targets with 6.0 wt% Ga_2O_3.

Acknowledgments

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References

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