

Low Resistive ZnO:Al Films with ZnO Buffer Layers Fabricated by Ar/N₂ Magnetron Sputtering

Ar/N₂マグネトロンスパッタによる低抵抗ZnO:Al膜の作製

Naho Itagaki^{1,2}, Kazunari Kuwahara¹, Kenta Nakahara¹, Daisuke Yamashita¹, Kunihiro Kamataki¹, Hyunwoong Seo¹, Giichiro Uchida¹, Kazunori Koga¹, Masaharu Shiratani¹, 板垣奈穂^{1,2}, 桑原和成¹, 中原賢太¹, 山下大輔¹, 鎌瀧晋礼¹, 徐鉉雄¹, 内田儀一郎¹, 古閑一憲¹, 白谷正治¹

¹Kyushu University, Motoooka 744, Fukuoka 819-0395, Japan.

²PRESTO, Japan Science and Technology Agency, Sanbancho 5, Chiyoda-ku, Tokyo 102-0075, Japan

¹九州大学システム情報科学研究所, 〒819-0395 福岡市元岡744

²JSTさきがけ, 〒102-0075 東京都千代田区3番町

Low resistive ZnO:Al (AZO) films with uniform spatial distribution have been obtained by utilizing buffer layers fabricated by Ar/N₂ magnetron sputtering. For 100 nm-thick AZO films, the averaged grain size of AZO films with buffer layers is 65 nm, which is 1.8 times larger than that of the films without buffer layers. This increase in the grain size of AZO films is due to the low grain density of buffer layers. As a result, the resistivity is drastically reduced. At the area facing the target erosion, the resistivity reduced from 2.27 mΩ•cm for the films without buffer layers to 0.50 mΩ•cm for our films, consequently, the spatial distribution of the resistivity is significantly improved. These results reveal that our method described here is full of promise for fabrication of ZnO-based TCO materials.

1. Introduction

ZnO is a fascinating II–IV semiconductor with unique features and a wide application range. The high electrical conductivity, high transparency to visible light, and material abundance make ZnO the most promising alternative to tin-doped indium oxide (ITO) for transparent conductive oxide (TCO). The resistivity of ZnO-based TCO, however, is higher than that of ITO and exhibits non-uniform spatial distribution. Since the crystallinity of ZnO films strongly affects both the carrier density and the mobility, it is important to control the crystallinity and its spatial distribution. Recently, we have developed a novel fabrication method of ZnO films utilizing solid phase crystallization (SPC) via Ar/N₂ sputtering deposition of amorphous zinc oxynitride (a-ZnON) films and subsequent annealing [1,2]. The resultant ZnO films have well aligned crystal orientation and higher crystallinity than the conventional sputtering films.

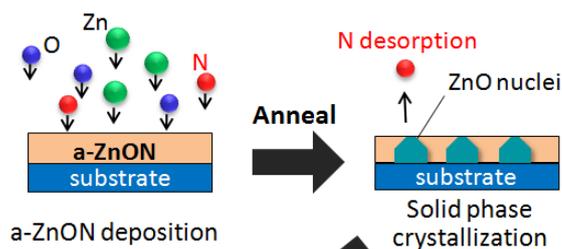
Here we utilize the SPC-ZnO films as buffer layers for ZnO:Al (AZO) TCO films and investigate their effects on the crystallinity of AZO films. Since the grain density of SPC films is lower than that of the conventional films, the buffer layers are expected to significantly improve the crystal grain size of AZO films. We also discuss the effects of buffer layers on the resistivity and its spatial distribution of AZO

films.

2. Experiment

Figure 1 shows the flow chart of the fabrication method for AZO films on the buffer layers prepared via SPC. The buffer layers were fabricated by annealing of a-ZnON films, which were deposited on quartz glass substrates by radio-frequency (RF) magnetron sputtering. The used targets were ZnO (2 inches in diameter). The sputtering gasses were

1. Fabrication of buffer layers via SPC



2. AZO deposition on SPC-seed layer

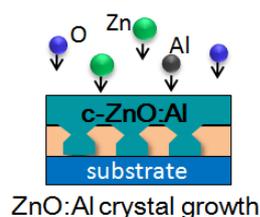


Fig. 1. Flow chart of the fabrication method for AZO films on the buffer layers prepared via SPC.

Ar and N₂. According to X-ray diffraction (XRD) analysis, the films were confirmed to be amorphous in nature. a-ZnON films were annealed at 200°C in an oxygen atmosphere and were converted to ZnO films. In the experiments, the thickness of seed layers was 10 nm, which was measured by spectroscopic ellipsometry and scanning electron microscope (SEM). AZO films were deposited on SPC buffer layers by RF magnetron sputtering with AZO targets (2wt% Al₂O₃). The used gas was Ar. The substrate temperature was kept at 200°C. No post annealing of AZO was performed. The crystal structures of the films were examined by X-ray diffraction (XRD) and transmission electron microscopy (TEM). The electrical properties of AZO films were evaluated by 4 point probe measurements and hall-effect measurements.

3. Results and discussion

Low resistive AZO films have been obtained by utilizing SPC buffer layers. Figure 2 shows the spatial distribution of the resistivity of AZO films (100 nm thick). The resistivity is drastically reduced, for instance, from 1.20 mΩ·cm to 0.39 mΩ·cm at the center (R = 0), which is attributed to improvement of crystallinity of AZO films. From the XRD measurements, it was observed that there is a significant difference in the intensity and the full width at half-maximum (FWHM) of ZnO (002) diffraction peak, that is, crystallinity and the crystallite size, between with and without SPC-buffer layers. TEM observation of the SPC seed layers shows that this increase in grain size is attributed to the low nuclei density of buffer layers. Furthermore, the fluctuation of the crystalline orientation of AZO film prepared on SPC-ZnO buffer layer is much smaller than that of the film without buffer layer.

The SPC buffer layers also improve the spatial distribution of the resistivity. For AZO films without buffer layers, the resistivity at the area facing the target erosion is 1.50 mΩ·cm, which is much higher than that at 60 mm away from the erosion (0.39 mΩ·cm). On the other hand, AZO films on SPC buffer layers exhibit uniform distribution of the resistivity of 0.51 - 0.25 mΩ·cm, respectively. These results reveal that controlling the crystallinity at the initial stage of AZO deposition is of great significance, especially at the area facing the target erosion where the energetic species impinging on the growing films can induce a high density of nucleation with broad orientation distributions.

From these results, we conclude that our method described here is full of promise for fabrication of

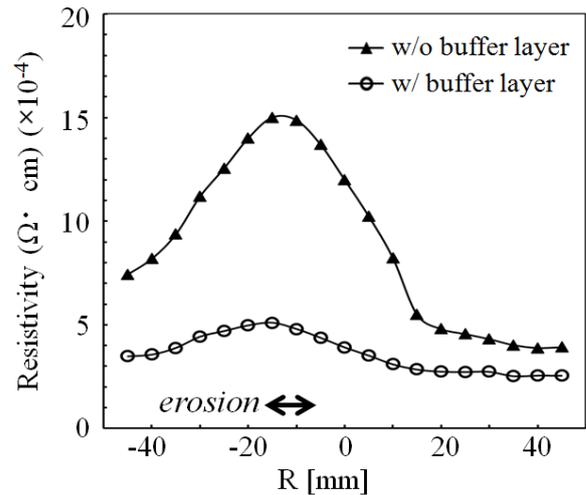


Fig. 2. Spatial distribution of the resistivity of 100-nm-thick AZO films with and without SPC buffer layers.

ZnO-based TCO materials.

3. Conclusions

The effects of SPC-ZnO buffer layers on the properties of ZnO-based TCO films have been studied. As a result, low resistive ZnO:Al (AZO) films have been obtained. The resistivity is drastically reduced from 1.50 mΩ·cm to 0.51 mΩ·cm at the area facing the sputtering target erosion, which is attributed to improvement of crystallinity of AZO films. The SPC buffer layers also improve the spatial distribution of the resistivity. From these results, we conclude that our method described here is full of promise for fabrication of ZnO-based TCO.

Acknowledgments

This work was partly supported by PRESTO from the Japan Science and Technology Agency.

References

- [1] N. Itagaki, K. Kuwahara, K. Nakahara, D. Yamashita, G. Uchida, K. Koga, and M. Shiratani, "Highly Conducting and Very Thin ZnO:Al Films with ZnO Buffer Layer Fabricated by Solid Phase Crystallization from Amorphous Phase", *Appl. Phys. Express* 4 (2011) 011101.
- [2] K. Kuwahara, N. Itagaki, K. Nakahara, D. Yamashita, G. Uchida, K. Kamataki, K. Koga, and M. Shiratani, "High quality epitaxial ZnO films grown on solid-phase crystallized buffer layers" to be published in *Thin Solid Films* (2011).