Identification of the state of deposition inert gas remaining in transparent conductive films deposited by magnetron sputtering Junichi NOMOTO^{1,*}

¹ Advanced Coating Technology Research Center, National Institute of Advanced Industrial Science and Technology (AIST), High Energy Research Organization, Tsukuba Central 5-2, 1-1-1 Higashi, Tsukuba, 305-8565, Japan

1 Introduction

Magnetron sputtering (MS) is most commonly deposition technique for transparent conductive oxide films. Magnetron-sputtered films, however, have an issue to be resolved: electrical resistivity (ρ) strongly depends on the substrate positions. ρ of the films grown at a position in the area of the substrates opposite to the erosion zone of the target is quite high compared with those of the films deposited at a position in the regions located far away from the above area. To date, dominant factors determining the above non-uniform of the spatial distribution of electrical properties are not yet thoroughly understood. We have been investigating the effects of the erosion zone of MS targets on the carrier transport of magnetron sputtered Aldoped ZnO (AZO) films prepared on a glass substrate to give a resolution to the issue described above. [1]

A series of the experimental results yielded the following. [1-3] For AZO films deposited by direct current (DC) MS, we found quite high amounts of residual argon (Ar) compared to AZO films deposited by radio frequency (RF) MS at any given substrate position; During film growth, recoiling Ar-related species attacked the AZO films grown at the substrate positions opposite to the erosion zone. Note that the impact of Ar atoms gives rise to the enhancement in the contribution of grain boundary scattering to the carrier transport owing to the poor crystallographic orientation between columnar grains. In addition, the residual Ar atoms present a significant obstacle to free carriers in ingrains, causing a decrease in intrinsic carrier mobility. [1-3] However, none of the previous studies offers the detailed information on the local structure of Ar in ZnO crystal.

In this work, a combination of Ar K-edge X-ray absorption spectroscopy (XAS) and first-principles calculations ware used to investigate the local structure of Ar in ZnO crystal.

2 Experiment

We deposited AZO films with systematic various Ar contents by 10 different deposition conditions. The substrate was alkali-free glass substrates (Corning, Eagle XG). The target was a high-density sintered ceramic circular AZO target (Toshima Manufacturing Corp.) with an Al₂O₃ content of 2.0 wt %. All of the deposition processes were performed under a pure Ar atmosphere at a pressure of 1.0 Pa. Prior to film deposition, the chamber was evacuated until the base pressure reached approximately 2.0×10^{-5} Pa. The typical film thickness of the obtained AZO films was approximately 500 nm.

The XAS measurements were performed on the beam line BL-11B at Photon Factory. The synchrotron radiation was monochromatized by a Si(111) double crystal monochromator. All the XAS experiments were carried out in a vacuum chamber at room temperature in the fluorescence yield mode using a Si drift detector. Incident X-rays were perpendicular to the sample surface and the detection angle was parallel to the sample surface to avoid detecting the scattered X-rays.

3 Results and Discussion

Figure 1 shows the typical Ar K-edge X-ray absorption near edge structure (XANES) spectrum for the AZO film. Three peaks at the energies of 3.20, 3.205 and 3.212 keV were clearly observed for all samples. The predominant peak at the energy of 3.20 keV corresponds to the localized transition of the 1s to 4p. [4] The peak at the photon energy of 3.212 keV is attributed to the cluster Ar Ka line. [4] These features are also observed from Ar K-edge XAS in DC sputtered aluminum film, which reported by Takemura *et al.*, [5] On the other hand, the peak at the energy of 3.205 keV has never been reported.



Fig. 1: Ar K-edge XANES spectrum for the AZO film

Acknowledgement

The authors express their sincere gratitude to Dr. Yoshinori Kitajima of High Energy Accelerator Research Organization for assistance with the XAS measurements.

References

- [1] J. Nomoto et al., J. Appl. Phys. 124, 065304 (2018).
- [2] J. Nomoto et al., ACS-Omega 4, 14526 (2019).
- [3] J. Nomoto *et al.*, *J. Vac. Sci. Technol. B* **38**, 022202 (2020).
- [4] E. Rühl et al., J. Chem. Phys. 98, 6820 (1993).
- [5] M. Takemura *et al.*, Spectrochim. Acta Part B, 54, 159 (1999).
- * nomoto.junichi@aist.go.jp