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Growth of V-doped ZnO Thin Films with Two-layer Structure on a-Al_2O_3 by RF Magnetron Sputtering

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Abstract

Drastic transition in c-axis orientation for V-doped ZnO (VZO) thin film grown on an a-face Al_2O_3 substrate was investigated. Through observations of TEM cross-sectional views, VZO films with two-layer structure were found. As a result of X-ray diffraction analyses changing the film thickness, the layer located immediately above the substrate was oriented along the c-axis direction, while the upper layer consisted of a mixture of c-axis and [10-11] direction. The initial thin layer grew coherently with compressive strain caused by V doping, and the extension of c-axis lattice constant was released gradually with thickening the VZO film up to about 200-nm thick. The crystal orientation tilted to [10-11] direction after the strain was fully relaxed. Conclusively, gradually relaxed thick layer affected crystal orientation of the upper layer.

1. Introduction

Transparent conductive oxides have been studied for many optoelectronic devices such as flat panel displays [1]. Doping of transition-metal ions, which take a wide variety of both valence binding configurations and coordination numbers, enables to achieve low resistivity, ferromagnetic property, and piezoelectricity [2]. In our previous work, anomalous dependence of crystallinity on V concentration in V-doped ZnO (VZO) thin films grown on a-Al_2O_3 substrate was observed [3]. That is, 500-nm-thick VZO films with increasing V up to about 1 at.% showed a sharp decrease in c-axis orientation and those with increasing V up to about 2 at.% recovered crystallinity, as shown in **Fig. 1**. This indicates that there is a possibility to control the crystal orientation by V concentration. Therefore, to investigate the effect of V doping on the film structure, crystal structure of VZO films with V of about 1 at.% depending on the film thickness was investigated.

2. Experiments

The VZO thin films were deposited by RF magnetron sputtering on a-face Al_2O_3 substrates using a ceramic ZnO target with V chips. Films of various thickness were deposited at substrate temperature of 450ºC under RF power of 150 W and Ar pressure of 1.0 Pa. Cross-sectional images of the films were obtained by TEM and crystal structures of VZO films were estimated by out-of-plane and in-plane XRDs.

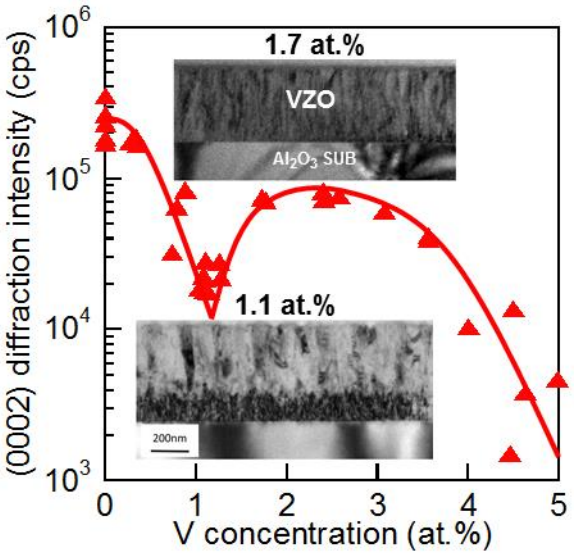

Fig. 1 Dependence of XRD (0002) diffraction peak intensity of 500-nm-thick VZO films on V concentration. Insets are TEM cross-sectional views of VZO films of 1.1 and 1.7 at.%.

3. Results

The TEM cross-sectional views of VZO films of 1.1 and 1.7 at.% are shown in the inset of Fig. 1. For 1.1-at.% VZO film, the film texture changed drastically at about 200-nm thick. On the other hand, for 1.7-at.% VZO film, it changed vaguely at about 50-nm thick.

Out-of-plane XRD patterns for 1.1-at.% VZO films of from 60- to 530-nm thick are shown in **Fig. 2**. For thin films of 60- and 125-nm thick, only the diffraction peak from (0002) plane was observed. However, the diffraction peak from (10-11) plane was also seen for the films over 215-nm thick. That is, the change of the film texture in the TEM image indicates the change of film structure with a mixture of two planes of (0002) and (10-11). Dependence of c- and a-axis lattice constants on film thickness was estimated from the diffraction angle from (0002) and (10-10) plane in the out-of-plane and in-plane XRD measurements (**Fig. 3**). As a result, the c-axis lattice constant decreased gradually and closed to about 5.205 Å (equal to the typical c-axis lattice constant of ZnO [4]). This can be considered that the coherent growth was performed at the initial stage of film deposition and the compressive strain was released with increasing film thickness. The film was fully relaxed

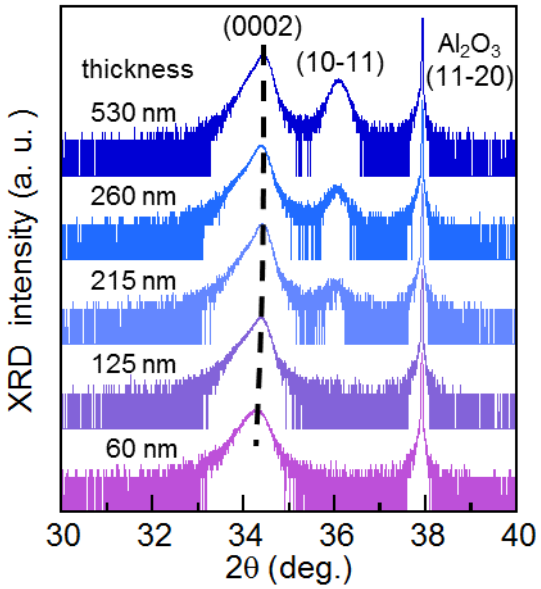

Fig. 2 Out-of-plane XRD patterns for 1.1-at.% VZO films of from 60- to 530-nm thick.

at about 200-nm thick and the crystal orientation tilted to [10-11] direction. Dependence of diffraction intensities from (0002) and (10-11) planes on film thickness is shown in **Fig. 4**. The increase of diffraction intensity from (0002) plane weakened for VZO film over about 200-nm thick, while the diffraction intensity from (10-11) plane increased proportionally to the thickness. This indicates that the film over about 200-nm thick grew mainly along [10-11] direction. As a result, VZO film with two-layer structure was formed. The films located immediately above the substrate and the upper film oriented along c-axis and [10-11] direction, respectively.

From these results, in the case of small V doping, gradually relaxed thick layer affected crystallinity of the upper layer and deteriorated the c-axis orientation. On the other hand, in the case of large V doping, the thin initial layer relaxed sharply and did not influence the c-axis orientation of the upper layer. In the meeting, the more details including grain size, crystal symmetry, and resistivity will be discussed.

4. Conclusions

Drastic transition in c-axis orientation for VZO thin film grown on an a-face Al_2O_3 substrate was investigated. From TEM cross-sectional views and X-ray diffraction analyses, VZO films consisted of two layers. The layer located immediately above the substrate was oriented along the c-axis direction, while crystal orientation of upper layer tilted to [10-11] direction. Conclusively, gradually relaxed thick layer affected crystal orientation of the upper layer.

Acknowledgement

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References

[1] K. Ellmer, Nat. Photonics **6** (2012) 809.
[2] N. A. Spaldin, Phys. Rev. B **69** (2004) 125201.
[3] H. Chiba et al., The 61st JSAP Autumn Meeting (2014) E10-8.
[4] Ü. Özgür et al., J. Appl. Phys **98** (2005) 041301.

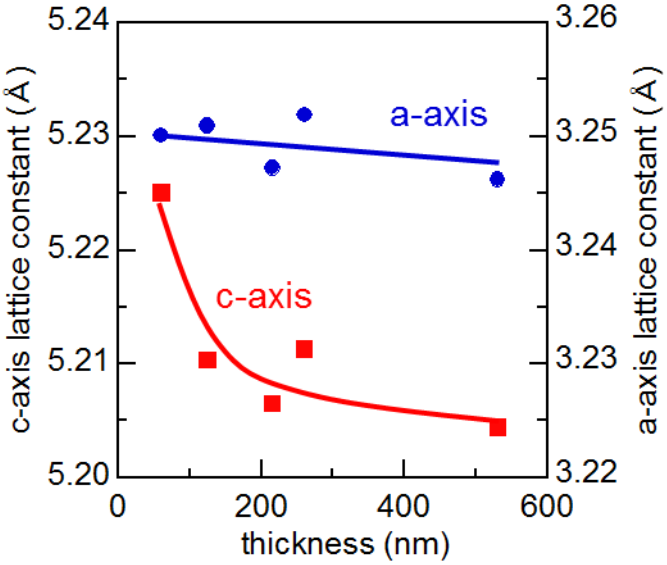

Fig. 3 Dependence of c- and a-axis lattice constant calculated by (0002) and (10-10) diffraction angle on the thickness for 1.1-at.% VZO films.

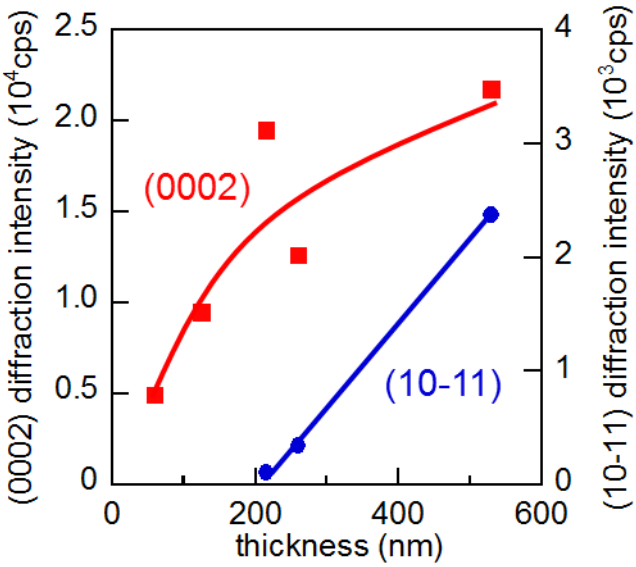

Fig. 4 Dependence of diffraction intensities from (0002) and (10-11) planes on the thickness for 1.1-at.% VZO films.