

# Surface modification of 10-nm thick ZnO films as interface layers to tailor the properties of Al-doped ZnO films and control of surface and interface roughness to achieve high carrier transport ultra-thin W-doped In<sub>2</sub>O<sub>3</sub> films

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## 1. Introduction

We have investigated factors limiting carrier transport of highly transparent conductive oxide (TCO) films such as ZnO- and In<sub>2</sub>O<sub>3</sub>-based films with thicknesses ( $t$ ) ranging from 5 nm to 2  $\mu$ m. To achieve high Hall mobility ( $\mu_{\text{H}}$ ) TCO films deposited on amorphous glass substrates, we have developed different types of nanoscale technology. In this talk, we elucidate the effect of the two types of technologies on  $\mu_{\text{H}}$  of TCO films deposited on the substrates.[1,2]

## 2. 10-nm-thick critical layer

The rf-magnetron-sputtered Al-doped ZnO (rf-MS-AZO) polycrystalline films have a texture with a well-defined (0001) orientation, whereas polycrystalline AZO films deposited by dc-MS (dc-MS-AZO) have a texture with a mixed orientation of the (0001) plane with other planes, as shown on the bottom right in Fig. 1, resulting in reduced  $\mu_{\text{H}}$  due to a high contribution of grain boundary scattering to carrier transport.

The analysis of the data obtained by atomic force microscopy (AFM:Park Systems NX10) measurements shows distinct differences in the surface roughness and structure between rf- and dc-MS-AZO films having  $t$  of more than 20 nm. We, thereby, develop a multistep deposition of dc-MS-AZO/10-nm-thick rf-MS-AZO/substrates [1]. Figure 1 shows cross-sectional schematic diagrams, AFM images and out-of-plane wide-range reciprocal space maps (RSMs) of the samples (RIGAKU, SmartLab), where  $R_{\text{a}}$  and  $R_{\text{q}}$  are average roughness and mean-square roughness, respectively. From Fig. 1, we concluded that the use of the 10-nm-thick critical layer produces improved surface roughness thick AZO films that have a texture with a well-defined (0001) orientation.

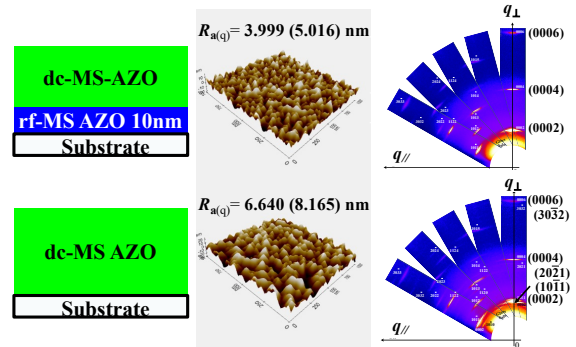


Fig. 1 cross-sectional schematic diagrams of samples, AFM images and out-of-plane wide-ranged reciprocal space maps of AZO films.

The results of Hall effect measurement at room temperature yield that the presence of the critical layers enhances the carrier concentration ( $n_e$ ) by 6.76 % as well as  $\mu_{\text{H}}$  by 15 % compared with AZO films deposited on critical-layer-free glass substrates. This results in a decrease in electrical resistivity ( $\rho$ ), that is inversely proportional to the products of  $n_e$  and  $\mu_{\text{H}}$ , by 18.3 % ( $\rho = 2.32 \times 10^{-4} \Omega\text{cm}$ ). Analysis of the experimental data clearly reveals that the increase of  $\mu_{\text{H}}$  is due to the fact that the GB scattering mechanism contributes very little to the carrier transport.

## 3. Specular or diffuse scattering

We achieve 5-nm-thick conductive polycrystalline W-doped In<sub>2</sub>O<sub>3</sub> ( $p$ -IWO) films showing  $\mu_{\text{H}}$  of 57.7  $\text{cm}^2/(\text{Vs})$  with solid-phase crystallization of amorphous IWO ( $a$ -IWO) films deposited on glass substrates (Coring EAGLE XG). We used reactive plasma deposition with dc arc discharge (RPD: Sumitomo Heavy Industries, Ltd.). The  $a$ -IWO films were deposited on the substrates without intentionally heating of them.

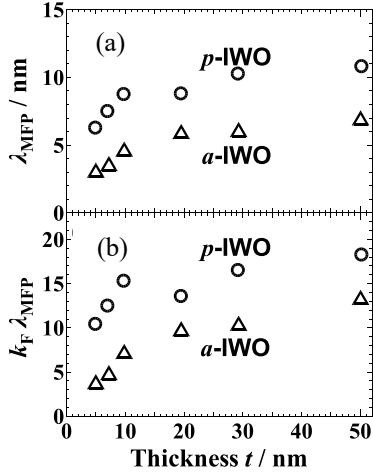


Fig. 2 (a) Mean Free path of carrier electrons ( $\lambda_{\text{MFP}}$ ) and (b) product of Fermi wave vector and  $\lambda_{\text{MFP}}$  of *a*-IWO films (triangles) and *p*-IWO films (circles) as functions of thickness ( $t$ ).

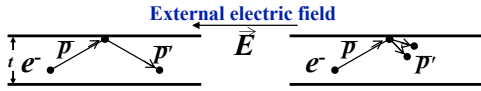


Fig. 3 (left) specular and (right) diffuse scattering

Figures 2(a) and (b) show the mean free path of carrier electrons ( $\lambda_{\text{MFP}}$ ) and (b) product of Fermi wave vector ( $k_{\text{F}}$ ) and  $\lambda_{\text{MFP}}$ , that is an important physical quantity which characterizes the degree of disorder in a conductor [2], of *a*-IWO films (triangles) and *p*-IWO films (circles) as functions of thickness ( $t$ ).  $\lambda_{\text{MFP}}$  are calculated using the experimental data,  $n_e$  and  $\mu_{\text{H}}$ , and  $k_{\text{F}}$  are obtained theoretically using  $n_e$ . From Fig. 2(a), for IWO films with  $t$  of less than 10nm, we find  $\lambda_{\text{MFP}}$  close to  $t$ . In such films, in case that dominant scattering mechanism at the film surface and film/substrate interface is not specular but diffuse scattering one (Fig. 3),  $\mu_{\text{H}}$  will be substantially reduced. We will discuss in more detail based on the classical size effect model below. Figure 2(b) shows that the condition for the metallic regime:  $k_{\text{F}} \times \lambda_{\text{MFP}} > 1$  is satisfied for all the *a*- and *p*-IWO films, in which the transport of charge carriers takes place via delocalized states. Note that Figure 2(b) clearly shows a abrupt decrease in the  $k_{\text{F}} \times \lambda_{\text{MFP}}$  with a decrease in  $t$  from 10 nm for *p*-IWO films. Here, however, we focus on the study about dominant factors limiting  $\mu_{\text{H}}$  based on the above classical size effect model.

In this work, as a resolution to the crucial issue, we optimized the energy of the flying particles, singly charged indium ( $\text{In}^+$ ) ions, to be 25 eV, to reduce the roughness of the surfaces and interfaces of less than 1 nm [2]. The aforementioned values of two different types of roughness has been confirmed by X-ray reflectivity (XRR) measurements.

In the ultrathin *p*-IWO films, we can distinguish three mechanisms of free-electron scattering: bulk, surface, and film/substrate interface ones. Each of these scattering mechanism has its characteristic relaxation time, and the resultant relaxation time of the films  $\tau_{\text{f}}$  is given as

$$\tau_{\text{f}}^{-1} = \tau_{\text{B}}^{-1} + \tau_{\text{sur}}^{-1} + \tau_{\text{if}}^{-1}, \quad (1)$$

where  $\tau_{\text{B}}$ ,  $\tau_{\text{sur}}$  and  $\tau_{\text{if}}$  denote the relaxation times of carrier electrons in the bulk, at the surface and interface scattering, respectively. The following conclusions were obtained from the comparison between the results of Hall effect measurements on the ultra-thin films of various thicknesses and the carrier mobility theoretically obtained using equation (1): When  $t$  is reduced from 10 nm, the scattering mechanism changes from specular to diffuse scattering. This results in reduced  $\mu_{\text{H}}$  with a decrease in  $t$ . One of the causes of the above scattering-mechanism change should be the localized distribution of oxygen atoms diffusely transferred to the surface and interface during postannealing, on the basis of analysis of the data obtained by high resolution Rutherford backscattering spectrometry (RBS) measurements. A solution to control the degree of this change is essential for widening TCO applications.

#### 4. Conclusion

The surface and/or film/substrate interface modification of TCO films with  $t$  of less than 10 nm is a very effective way for tailoring carrier transport of TCO films.

#### References

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