DOMINANT ROLE OF GRAIN BOUNDARY SCATTERING ON CARRIER TRANSPORT OF HIGHLY TRANSPARENT CONDUCTIVE Ga-DOPED ZnO FILMS

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Introduction

Transparent conducting electrodes (TCEs) are a necessary component of all types of flat display panels (FDPs), such as liquid crystal display panels. The TCEs must exhibit high conductivity and high optical transmittance. Indium tin oxide (ITO) films are widely used as TCEs in various types of FDPs. Recently, as an alternative to using ITO in FDPs, a great deal of interest has been focused on highly TCEs made from zinc oxide (ZnO), which has similar electrical and optical properties to ITO [1], because of its advantages over ITO, such as low material cost and non-toxicity.

In our previous works [4-6], we reported the dependence of electrical resistivity ρ on the thickness (d) of GZO films: ρ decreases with increasing d. Note that the d dependence of ρ includes variations with both the grain size and the alignment of the c-axis between the columnar grains in polycrystalline GZO films.

In this study, we performed a systematic study of the d dependence of the structural and electrical properties of GZO films. The aim of this study is to investigate the effects of the grain boundary (GB) scattering on the carrier transport of GZO films in order to clarify the issues to be resolved in realizing a wide variety of applications.

Experimental

GZO films with different ds ranging from 100 to 500nm were deposited on alumino silicate glass substrates (HOYA NA32R) at a T_s of 200°C with a deposition rate of 170 nm/min by ion-plating deposition with DC arc discharge [2-4]. The evaporation source was sintered ceramic ZnO (99.99% purity) containing 4wt% Ga_2O_3 (99.9% purity, HAKUSUI Tech. SKY-Z). The base pressure and deposition pressure were 5.0×10^{-5}Pa and 0.22Pa, respectively.

The structural analysis was carried out using an X-ray diffraction (XRD) measurement system (Cu $K\alpha 1$, wavelength λ=0.1540562nm) (Rigaku ATX-G). ρ, carrier concentration N_e and Hall mobility μ_H of films with different ds were determined by Hall effect measurement using the van der Pauw method (Nanometrics, HL5500PC). The optical mobility μ_{opt} was estimated by multi angle spectroscopic ellipsometry (J. A. Woollam, M-2000DI) combined with an appropriate model of the raw data: the Drude model combined with the Tauc-Lorentz model [4].

Results and Discussion

Structural Properties

The d dependence of the crystal structure of the GZO films was investigated by high-resolution XRD measurements. Both out-of-plane and in-plane XRD patterns show that polycrystalline GZO films of all d have a wurtzite structure with strong c-axis orientation perpendicular to the substrate. No other phase, such as that of Ga_2O_3 or Zn metal, was detected in the XRD analysis.

Fig. 1 shows both the full width at half maximum (FWHMω) of the (0002) rocking curve and the lateral size of the grains (hereafter referred to as the grain size, L), estimated by analyzing data obtained from the in-plane XRD pattern, in the direction parallel to the substrate plane as a function of d. With increasing d, the FWHMω of the GZO films rapidly decreases up to a thickness of 200nm, followed by a gradual decrease.

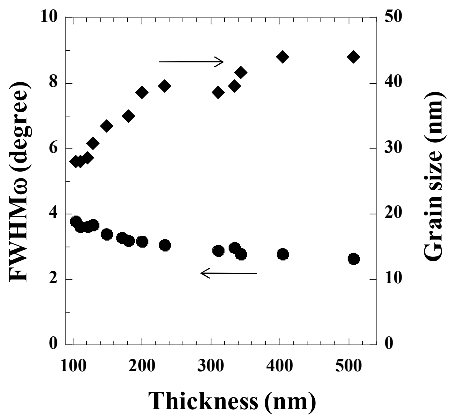
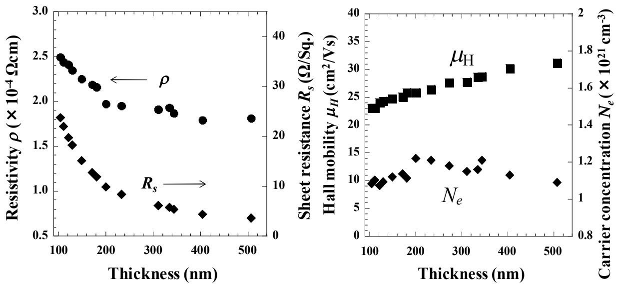


Fig. 1 Full width at half maximum (FWHMω) of the (0002) rocking curve and the lateral size of the grains in the direction parallel to the substrate plane as a function of thickness of GZO film.

This shows that the alignment of the *c*-axis between the columnar grains in polycrystalline GZO films is improved as *d* increases. On the other hand, *L* sharply increases up to a *d* of 200nm, above which it changes little up to 350nm, after which it increases gradually.

Electrical Properties

Fig. 2 shows (a) the electrical sheet resistance R_s and ρ and (b) N_e and μ_H of the GZO films as a function of *d*. ρ decreases as *d* increases: ρ (R_s) has a minimum of 1.8×10^{-4} Ωcm (3.57Ω/Sq.) at 404nm (507nm). Note that (1) the *d*-dependent behaviour of ρ was very similar to that of FWHMω, as shown in Fig. 1.: with increasing *d*, ρ decreases sharply up to 200nm, above which it decreases gradually and (2) Fig. 2(b) shows that the *d*-dependent behavior of ρ is mainly due to that of μ_H.



(a) (b)

Fig. 2 (a) Resistivity ρ and s heet resistance R_s and (b) Hall mobility μ_H and Carrier concentration N_e of GZO films as a function of thickness.

Effect of GB scattering on carrier mobility

Grain boundaries in polycrystalline films disturb the translational symmetry of the crystal and inevitably cause the scattering of free carriers.

Fig. 3 shows μ_{opt} and μ_H as a function of *d* for the GZO films. For all samples, it shows that μ_{opt} remains almost constant at an average value of 28.6cm^2/Vs. When μ_H is limited by the two phases of in-grain and GB, it can be expressed as [4],

$$\frac{\mu_{opt}}{\mu_{gb}} = \frac{\mu_{opt} - \mu_H}{\mu_H} \qquad (1)$$

where μ_{gb} indicates carrier mobility at GB.

For a 105-nm-thick GZO film with a grain size of 28nm, the value of μ_{opt}/μ_{gb} is 0.21, whereas for a 200-nm-thick GZO film with a grain size of 39nm, it is 0.11. With further increasing *d*, μ_H approaches μ_{opt} as shown in Fig. 3. For a 344-nm-thick GZO film with a grain size of 42nm, μ_{opt}/μ_{gb}~0, and we obtain a very low ρ of 1.87Ωcm.

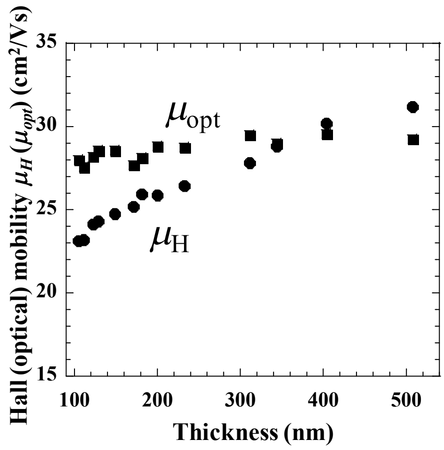


Fig. 3 Optical mobility μ_{opt} and Hall mobility μ_H as a function of thickness of GZO films.

Conclusion

The experimental study of a continuous transition from GB scattering to ingrain-scattering limited carrier transport within highly transparent conductive GZO polycrystalline films with *d* is reported. This study indicates that carrier transport trough GB can be improved by increasing the grain size and improving the alignment of the *c*-axis between the columnar grains in polycrystalline GZO films with (0001)-oriented columnar structure.

Acknowledgement

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