Transactions of the Materials Research Society of Japan 25 [1] 341-344 (2000)

Deposition of Well Oriented Polycrystalline ITO Films on ZnO-Coated Polymer Substrates by dc Sputtering

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ITO films were deposited by dc sputtering on glass, acrylic-coated polycarbonate (PC), ZnO-coated glass (ZnO/glass) or ZnO-coated PC (ZnO/PC) substrates. These ZnO underlayers deposited also by dc sputtering showed <001>-preferred orientation in the wide range of the deposition conditions and were used as bufferlayers to control crystallographic orientation of the ITO films at the initial stages of the growing films. The XRD analysis revealed tha ITO films deposited on glass (ITO/glass) and PC (ITO/PC) substrates with ITO thicknesses thinner than about 150 nm were amorphous, whereas ITO films deposited on ZnO/glass (ITO/ZnO/glass) and ZnO/PC (ITO/ZnO/PC) showe polycrystalline structure with predominant <111>-preferred orientation, indicating heteroepitaxial growth of In_2O_3 (111)||ZnO (001) relation. The thicknesses of ZnO underlayer necessary for the ITO heteroepitaxial growth wer confirmed to be more than 5 nm (nominal value) for the glass and PC.

Key words: ITO, ZnO, dc sputtering, heteroepitaxial, polycarbonate

1. INTRODUCTION

Sn-doped In_2O_3 (ITO) films have many applications such as transparent electrodes for solar cells$^{1-3)}$, liquid crystal displays (LCDs), or other flat panel display devices (FPDs)$^{4-6)}$, because they have high conductivity and high transparency in the visible region of light. There is a strong current commercial requirement that the high quality ITO films for color LCDs or touch panel-type “flexible” LCDs should be deposited at low substrate temperature (Ts: RT~150°C)on organic color filters or flexible polymer substrates such as polycarbonate (PC) or polyethylene terephthalate (PET) whose thermostability in vacuum is restricted. Generally, polycrystalline structure of ITO films is affected by a kind of substrate such as amorphous, polycrystalline and single crystal. Despite that, there have been relatively few studies for a correlation between the growth mechanism of ITO films and the structures of underlayers during initial stages of the film growth. Recently, it has been reported that ITO films deposited on <001>-preferred oriented polycrystalline ZnO films showed predominant <111> orientation.$^{7)}$

In this study, polycrystalline ITO films with the high degree surface normal orientation (not the in-plane orientation) have been successfully deposited on soda-lime glass and polymer substrates by using ZnO polycrystalline films as bufferlayers between ITO and the substrates. The correlation between microstructure and surface morphologies of the ITO films deposited with different thicknesses of ZnO underlayers were investigated.

2. EXPERIMENTAL

ITO films were deposited on various substrates [glass (ITO/glass) or acrylic-coated PC (ITO/PC)] or ZnO-coated substrates (ITO/ZnO/glass or ITO/ZnO/PC) by a dc magnetron sputtering using a high-density ceramic ITO target (SnO_2: 10 wt%, Tosoh Co., Ltd, packing density: more than 99.9 %). The ZnO underlayers with various thicknesses (about 5 ~ 320 nm) were deposited on soda-lime glass or acrylic coated PC substrates also by the dc magnetron sputtering using a ceramic ZnO target (packing density: about 95 %), which was known to exhibit the self-texture of <001>-preferred orientation (c-axis is normal to the substrate surface) in a wide range of deposition conditions. All the sputter depositions were carried out using Ar gas only without substrate heating (Ts<50°C). The distance between the target and substrates was 100 mm and dc sputtering power was 50 W. The ITO and ZnO underlayers were deposited at gas pressures of 0.7 Pa and 0.5 Pa, respectively.

The film thickness was measured with a Dektak3 surface profiler (Sloan Tech.). The nominal thickness was estimated by the linear fitting of the deposition time for the thinner films (~10 nm). The X-ray diffraction (XRD) was carried out with 40kV, 20 mA CuKα1 radiation by θ-2θ mode. Surface morphology of the ITO films was observed using atomic force microscopy (AFM, Shimazu SPM-9500). Room temperature resistivity, Hall mobility and free carrier density of the ITO films were measured by the four-point probe method and Hall-effect measurement in the van der Pauw geometry (Bio-Rad HL-550PC).

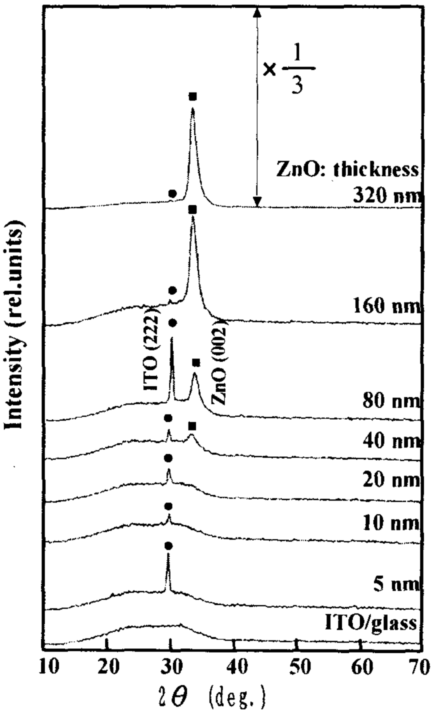

Fig. 1. X-ray diffraction patterns of the ITO films (about 150 nm) deposited on glass substrate (ITO/glass) and ZnO-coated (about 5~320 nm) glass substrate (ITO/ZnO/glass).

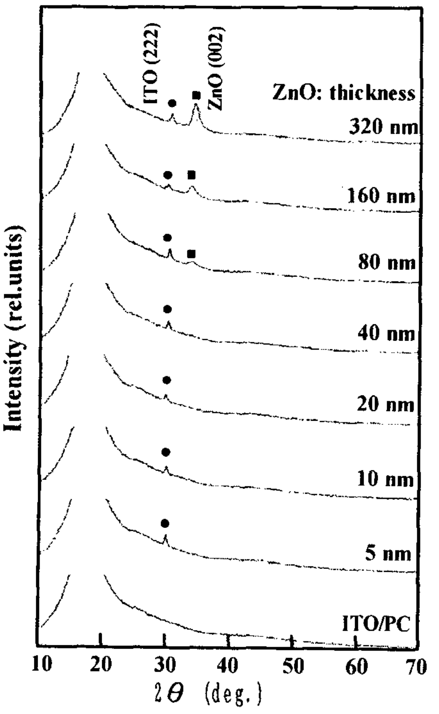

Fig. 2. X-ray diffraction patterns of the ITO films (about 150 nm) deposited on PC substrate (ITO/PC) and ZnO-coated (about 5~320 nm) glass substrate (ITO/ZnO/PC).

3. RESULTS AND DISCUSSION

Figure 1 shows XRD patterns of ITO (about 150 nm) films deposited on glass substrate (ITO/glass) or ZnO-coated glass (ITO/ZnO/glass) with different ZnO thickness (about 5~320 nm). In the case of ITO/glass, XRD profiles of the films showed only broad pattern indicating that the films were entirely amorphous structure. On the other hand, in the case of ITO/ZnO/glass, ITO films, even at thickness of ZnO underlayer of 5 nm (nominal thickness), showed clear <111>-preferred orientation. The average neighboring oxygen-oxygen (O-O) distances weighted by number/unit for the In_2O_3 lattice plane parallel to (111) was calculated to be 0.33534 nm 7), whereas the O-O distance on the ZnO lattice planes parallel to (001) is 0.32498 nm (same as the lattice constant, a_{ZnO}), where the oxygen atoms are located at apexes of regular equilateral triangles. Therefore, a mismatch of the O-O distance between $(111)_{In_2O_3}$ and $(001)_{ZnO}$ could be estimated to be only 3 %, which should be the reason for the (111)-preferred orientation and improved crystallinity for ITO/ZnO/glass. An apparent improvement in crystallinity of ITO films was not observed with increasing film thickness of ZnO.

Figure 2 shows XRD patterns of ITO films deposited on PC (ITO/PC) and ZnO-coated PC (ITO/ZnO/PC) substrates. The polycrystalline heteroepitaxial ITO films were also successfully deposited on PC substrates by using preferred oriented ZnO films as bufferlayers.

Figures 3 and 4 show AFM images of the ZnO/glass and ZnO/PC [thickness of ZnO films: (a) 0 nm, (b) 20 nm, (c) 80 nm, (d) 320 nm]. These surface roughnesses were quantified by the average roughness (Ra), where the more gentle roughness of its surface was ejected using high pass filter for ZnO/PC because the surface of the bare PC substrate showed the characteristic surface morphology with gentle wave. As shown in Fig. 3, the surface of the bare glass substrate was smooth [Ra < 0.2 nm], whereas the surface of ZnO/glass became rough from Ra=0.2 to 7.5 nm with increasing film thickness of ZnO from 0 to 320 nm. Ra of ZnO/PC also increased with increasing film thickness of ZnO (Fig. 4). The increasing surface roughness of ZnO films is considered to be caused by the growth in crystal grains of ZnO

Figures 5 and 6 show AFM images of ITO films (about 150 nm) deposited on ZnO/glass (Fig. 3) or ZnO/PC (Fig. 4), where the thicknesses of the ZnO underlayers were (a) 0 nm, (b) 20 nm, (c) 80 nm, (d) 320 nm. The surface of the ITO films directly deposited on glass substrate was relatively smooth [Ra: 0.6 nm], whereas the surface of ITO/ZnO/glass became rough [Ra: 0.8 ~ 3.6 nm] with increasing film thickness of ZnO underlayer. The same tendency was observed in the PC substrate, compared with the glass substrate. The smooth surface morphologies of ITO/glass or ITO/PC are considered to be corresponds to the amorphous ITO films. On the other hand, these rough surface morphologies of ITO/ZnO/glass and ITO/ZnO/PC were in good agreement with the results of ZnO/glass (see Fig 3) and ZnO/PC (see Fig. 4). From results of Figs. 1 and 2, the crystallinity of ITO films was not appreciably affected by thickness of ZnO underlayer, implying that

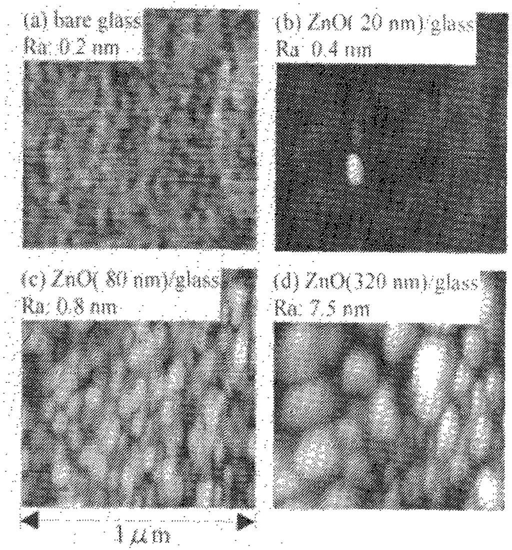

Fig. 3. AFM images of bare glass and ZnO(20~320 nm)/glass.

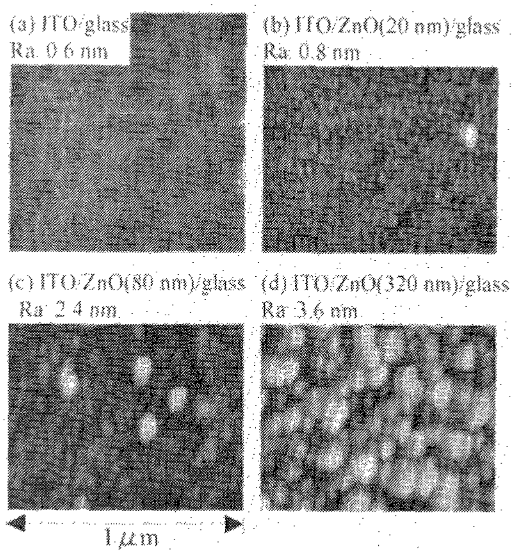

Fig. 5. AFM images of ITO/glass and ITO/ZnO(20~320 nm)/glass.

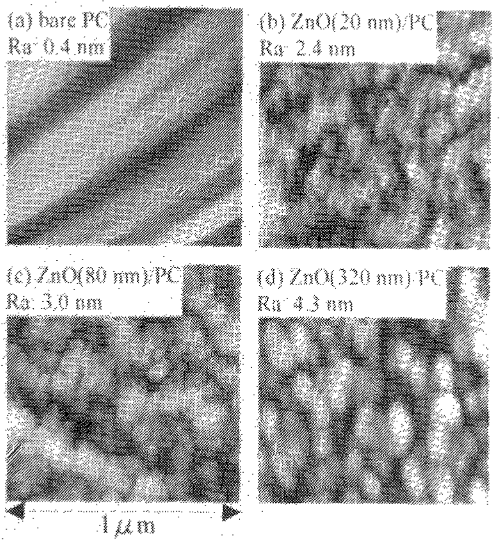

Fig. 4. AFM images of bare glass and ZnO(20~320 nm)/glass.

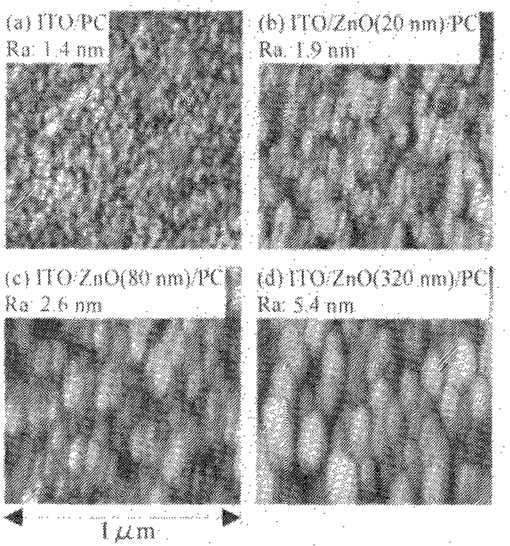

Fig. 6. AFM images of ITO/PC and ITO/ZnO(20~320 nm)/PC.

Table I The room temperature resistivity ρ, Hall mobility μ, carrier density n of ITO/glass, ITO/PC, ITO/ZnO/glass and ITO/ZnO/PC.
The thickness of ZnO and ITO films were about 5 and 100 nm, respectively.

	Resistivity (ρ) ($\times 10^{-3}\,\Omega\cdot$cm)	Mobility (μ) (cm^2/V$\cdot$s)	Carrier density (n) ($\times 10^{20}$/cm^3)
ITO/glass	0.81	26.9	2.61
ITO/PC	1	26.4	2.33
ITO/ZnO/glass	1.2	21.8	2.38
ITO/ZnO/PC	0.97	27.1	2.46

the surface morphologies of ITO/ZnO/glass or ITO/ZnO/PC were mainly affected by the surface morphology of ZnO underlayers.

The room-temperature resistivity ρ, Hall mobility μ, and carrier density n of ITO (about 100 nm) films deposited on various substrates [ITO/glass, ITO/PC, ITO/ZnO (nominal thickness: 5 nm) /glass and ITO/ZnO (nominal thickness: 5 nm) /PC] are shown in Table I. ITO/PC showed higher ρ ($1.0 \times 10^{-3}\ \Omega$ cm), compared with ITO/glass ($0.8 \times 10^{-3}\ \Omega$ cm), which should due to the low crystallinity caused by degassing from the PC substrate during initial film growth processes. The improvements in electrical properties were not clearly observed by using the ZnO underlayers.

4. CONCLUSION

The predominately <111> oriented polycrystalline ITO films were deposited on the <001> preferred oriented polycrystalline ZnO-coated glass or PC substrates at RT by dc magnetron sputtering without substrate heating. The ITO films deposited on glass or PC substrates were amorphous structure, whereas in the case of using ZnO underlayer with nominal thickness of 5 nm, XRD patterns of the ITO films showed <111>-preferred orientation. AFM images showed that the surface morphology of ITO/ZnO/glass or ITO/ZnO/PC was mainly dominated by that of ZnO films, where a clear change in electric properties by using ZnO underlayer was not observed.

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(Received December 17, 1999; accepted March 11, 2000)