Vacuum 86 (2011) 443–447

Contents lists available at SciVerse ScienceDirect

Vacuum

journal homepage: www.elsevier.com/locate/vacuum

Effect of SiO_2 buffer layer thickness on the properties of ITO/Cu/ITO multilayer films deposited on polyethylene terephthalate substrates

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A R T I C L E I N F O

Article history:
Received 24 March 2011
Received in revised form
13 September 2011
Accepted 13 September 2011

Keywords:
Indium tin oxide
Copper
SiO_2 buffer layer
Flexible substrate

A B S T R A C T

Transparent conductive ITO/Cu/ITO films were deposited on polyethylene terephthalate (PET) substrates with a SiO_2 buffer layer by magnetron sputtering using three cathodes at room temperature. The effect of the SiO_2 buffer layer thickness on the electrical and optical properties of ITO/Cu/ITO films was investigated. The ITO/Cu/ITO film deposited on the 40 nm thick SiO_2 buffer layer exhibits a sheet resistance of 143Ω/sq and transmittance of 65% at 550 nm wavelength. Highly transparent ITO/Cu/ITO films with a transmittance of 80% and a sheet resistance of 98.7Ω/sq have been obtained by applying −60 V substrate bias.

1. Introduction

Indium tin oxide (ITO) transparent conductive films have been used as transparent electrodes in organic light emitting diodes, liquid crystal displays, plasma display panels and solar cells [1–3]. Deposition of ITO films on glass substrates has been widely investigated during last decade. Recently, there has been considerable interest in the use of ITO films deposited on polymer substrates because polymer substrates are suitable for flexible display devices and electronics. Polymers are lightweight and flexible, but they easily absorb moisture and gas, have a lower thermal resistance, a weaker mechanical strength and a higher thermal expansion coefficient [4–7]. ITO films are usually deposited at a temperature higher than 250 °C and annealed at a temperature higher than 300 °C in order to obtain high transmittance and conductivity [8]. High temperature is not an effective process condition when ITO films are deposited on a polymer substrate to be applied in flexible optoelectronic devices. One way to improve the optoelectronic properties of the ITO films without substrate heating or post-annealing is to use ITO/Cu/ITO sandwich structure films which have lower resistivity than ITO single layer films of the same thickness [9–12]. A SiO_2 buffer layer is inserted between the polymer substrate and ITO/Cu/ITO films to reduce diffusion of moisture and gas. In this work, the effect of SiO_2 buffer layer thickness on the properties of ITO/Cu/ITO films was investigated.

2. Experimental

The SiO_2 buffer layers with the thickness ranging from 0 to 80 nm were deposited on PET (125 μm thickness) substrates by radio frequency magnetron sputtering SiO_2 ceramic targets (purity of 99.99%). ITO/Cu/ITO multilayer films were deposited on the top of SiO_2 buffer layer by direct current magnetron sputtering ITO targets (purity of 99.99%, In_2O_3:SnO_2 = 90:10 wt.%) and Cu targets (purity of 99.999%). Sputtering was performed at room temperature in an argon atmosphere with a target-to-substrate distance of 60 mm. The experimental deposition conditions are listed in Table 1. The thickness of each layer in the ITO/Cu/ITO films was kept constant at 40 nm/5 nm/35 nm.

Optical transmission was measured using a double beam spectrophotometer (TU1901). The resistivity ρ, free carrier concentration n, and Hall mobility μ_H were determined from Hall Effect measurements using the Vander Pauw method (Accent HL5500 Hall System) at a constant magnetic field of 0.517T. The structural properties of the films were determined using X-ray diffraction measurements with Cu-Kα radiation (Y-2000). The surface morphology of the films was examined by atomic force microscope (AFM).

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doi:10.1016/j.vacuum.2011.09.005

Table 1
Deposition conditions of SiO_2, ITO and Cu thin films.

	SiO_2	ITO	Cu
Base pressure (pa)	6×10^{-4}	6×10^{-4}	6×10^{-4}
Deposition pressure (pa)	0.5	0.51	0.3
Power density (w/cm^2)	RF, 2.83	DC, 1.66	DC, 0.92
Deposition rate (nm/min)	2.02	29.2	15.28
Gas flow rate (Ar sccm)	20	20	20

3. Results and discussion

Fig. 1 shows the XRD patterns of ITO/Cu/ITO films deposited on PET substrates with SiO_2 buffer layers ranging from 0 to 80 nm and bare PET. The peaks appearing at 26° and 54° indicate the PET substrate. No other distinct peak is found, which indicates that the ITO/Cu/ITO films with different thicknesses of SiO_2 buffer layers are all amorphous in structure.

Surface morphologies of the ITO/Cu/ITO films with buffer layers were investigated by AFM. Fig. 2 shows pictures of bare PET and ITO/Cu/ITO films with SiO_2 buffer layers. The surface of the ITO/Cu/ITO films with buffer layers is smoother. Fig. 2(d) shows that SiO_2(40 nm)/ITO/Cu/ITO film has the smoothest surface, which indicates that the surface roughness of the ITO/Cu/ITO films is strongly affected by the underlying SiO_2 buffer layers. First of all, substrate usually provides the template for subsequent deposition and growth of SiO_2 films. Atoms grow preferably in sunken regions on the substrate. Moisture and gas from the PET substrate diffuse into the SiO_2 films, which results in porous films. The SiO_2 layer becomes denser with depositing thickness, a SiO_2 layer with 40 nm thickness being sufficient to prevent degassing from the PET substrate. As the SiO_2 film continues to increase, the SiO_2 film shows a disorder distribution due to the full stress relaxation, resulting in a deterioration of the ITO/Cu/ITO film. The net effect is that the ITO/Cu/ITO film with a 40 nm SiO_2 buffer layer has the smoothest surface.

Fig. 3 shows the transmittance of the ITO/Cu/ITO films with different SiO_2 buffer layers in the wavelength range of 300–800 nm, with air as the reference. The PET substrate has a 90% transmittance at 550 nm, and the ITO/Cu/ITO films with SiO_2 buffer layers deposited on PET have a transmittance of about 63 ~ 65% at 550 nm. The ITO/Cu/ITO film without a SiO_2 buffer layer has higher transmittance in the ultraviolet region but almost the same transmittance as the films with SiO_2 buffer layers in the visible region, which indicates that the optical properties mainly depend on the Cu layer [13], SiO_2 buffer layers have little effect on the transmittance of all the ITO/Cu/ITO films.

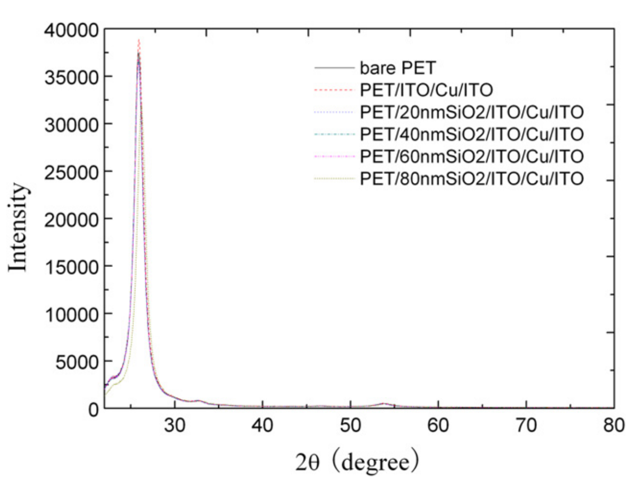

Fig. 1. XRD patterns of ITO/Cu/ITO films deposited on PET substrates with different thickness of SiO_2 buffer layers.

Fig. 4(a) shows the sheet resistance and resistivity of ITO/Cu/ITO films with SiO_2 buffer layers of different thickness. As is well known, PET is very sensitive to moisture and gas, so that a PET substrate will absorb and permeate substantially more moisture and gas than a glass substrate [14]. During deposition, these contaminants would flow out and diffuse into the films, which would work as acceptors or scattering centers and adversely affect their electrical properties. From Fig. 4(a) it can be seen that the overall sheet resistance and resistivity of ITO/Cu/ITO films with SiO_2 buffer layers is less than that of ITO/Cu/ITO films on bare PET substrates. The decrease in sheet resistance with an optimized buffer layer is believed to be due to the combined effect of surface smoothing and prevention of moisture and gas diffusion into the ITO/Cu/ITO films.

The total resistance of a multilayer is a combination of the resistances of three consecutive layers in parallel as follows: $1/R_{Total} = 1/R_{ITO(Bottom)} + 1/R_{Cu} + 1/R_{ITO(Upper)}$. Since R_{Cu} is much less than other resistances, the total resistance R_{Total} would mainly depend on the Cu layer. The metal resistance originates from the collision of free electrons which results in the loss of the directional speed accelerated from the electric field. Collisions may take place in electron-lattice, electron-impurity, electron-grain boundary and electron-surface. For a 5 nm thick Cu film, the resistivity is dominated by electron-surface collisions. The electron-surface collisions become severe and resistivity of the Cu film increases as the roughness of surface increases. A smoother surface has a positive effect on decreasing resistivity. A comparison of the Ra in Fig. 2 shows that ITO/Cu/ITO films with a 40 nm SiO_2 buffer layer have the smoothest Cu interlayer. The sheet resistance and resistivity are related to the change of ITO/Cu/ITO surface morphology.

Fig. 4(b) shows the dependence of the Hall mobility and carrier concentration of the ITO/Cu/ITO films on the SiO_2 buffer layer thickness. The Hall mobility of the ITO/Cu/ITO films decreases with increasing SiO_2 buffer layer thickness. As the SiO_2 buffer layer thickness increases, the carrier concentration of ITO/Cu/ITO films increases at first and then decreases slowly, finally increasing steeply. The resistivity of the ITO/Cu/ITO film decreases with increasing SiO_2 buffer layer thickness ($\leq$40 nm) due to an increase in carrier concentration. The Hall mobility of the ITO/Cu/ITO film is mainly related to the crystallinity, the defects and the surface properties. The Hall mobility of the ITO/Cu/ITO film decreases due to the deteriorated crystallinity caused by stress with increasing SiO_2 buffer layer thickness. The charge carriers of the ITO/Cu/ITO film consist of the free electrons of the Cu film, the electrons provided by the oxygen vacancies and the substitution of Sn^{4+} for In^{3+} in ITO films, the hole provided by the substitution of Cu^{2+} for In^{3+} in the diffusion interface of ITO/Cu. The variation of SiO_2 buffer layer has an effect on the structure of ITO/Cu/ITO multilayer, which results in the variation of the carrier concentration. The ITO/Cu/ITO film with a 40 nm SiO_2 buffer layer shows a minimum resistivity, which indicates that introduction of a SiO_2 buffer layer with proper thickness can lower the resistivity of the ITO/Cu/ITO films [7].

Fig. 5 shows the figure of merits (φ_{TC}) of ITO/Cu/ITO films with different SiO_2 buffer layers, and it is an important index for evaluating the performance of transparent conducting oxide (TCO) films. The φ_{TC} is defined as $\varphi_{TC} = T^{10}/R_s$, where T is the optical transmittance (at 550 nm in this study) and R_s is the sheet resistance [15]. The φ_{TC} reaches a maximum of $9.4 \times 10^{-5}\Omega^{-1}$ for the ITO/Cu/ITO film with a 40 nm SiO_2 buffer layer, which is higher than the φ_{TC}, $3.8 \times 10^{-5}\Omega^{-1}$, of the ITO/Cu/ITO film without