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LETTER

Enhancement of the electrical properties of Al-doped ZnO films deposited on ZnO-buffered glass substrates by using an ultrathin aluminum underlayer

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Transparent conducting oxide (TCO) films have been widely used as optoelectronic devices such as touch panels, flat panel displays (FPD), and thin film solar cells [1–5]. For display applications with high quality, the TCO films should have high optical transmittance in the visible region and high electrical conductivity. Indium tin oxide (ITO) has been a predominant TCO used for many years mainly for the flat panel display industry [6, 7]. However, impurity-doped zinc oxides (ZnOs) are widely accepted as substitutes for ITO-based TCO because of the advantages of low cost, resource availability (about a factor of 1,000 more abundant than indium), nontoxicity, and high thermal/chemical stability. Undoped ZnO usually presents a high resistivity due to a lower carrier concentration. Enhancement of the electrical properties of TCOs, specifically conductivity, can be achieved by increasing either the carrier concentration or the carrier mobility. Aluminum (Al), indium (In), and gallium (Ga) have been reported as effective dopants for zinc oxide-based TCO films [8–11]. Among the ZnO films doped with these elements Al-doped zinc oxide (AZO) films show the lowest electrical resistivity, which is close to that of ITO films. AZO films are also wide band gap semiconductors (E_g = 3.4–3.7 eV), which have high optical transmittance in the visible wavelength region [12]. However, a further decrease in resistivity is required for future display technology applications. Recently using an ultrathin silver (Ag) interlayer has been reported to enhance the conductivity of TCO films [13–15]. It is, however, known that the transmittance of the multilayer TCO films decreases significantly with an increase in the Ag interlayer thickness although the electrical properties are enhanced.

Various deposition methods such as thermal evaporation, chemical vapor deposition, spray pyrolysis, sol–gel process, pulsed laser deposition, and sputtering have been used for deposition of AZO and metal films. Among these techniques sputtering is known to be the most favorable deposition method to obtain highly uniform films at a high deposition rate [16]. In this article, the electrical and optical properties of the AZO/Al multilayer film deposited on ZnO-buffered glass by using a radio frequency (RF) magnetron sputtering technique are presented.

The AZO/Al multilayer films were deposited on ZnO-buffered glass by sequencial RF magnetron sputtering of ZnO, Al, and AZO targets on alkali-free glass substrates. The thicknesses of the AZO layer and the ZnO buffer layer were fixed at 200 and 50 nm, respectively, and the Al layer thickness was varied in a range of 0–20 nm with an increment of 5 nm. An RF sputtering system with a two-stage turbomolecular pumped chamber was used. AZO and ZnO sputter targets with 99.9% purity were used. All depositions were carried out at room temperature. The sputter chamber was equipped with a load lock system, and the base pressure of the sputter chamber prior to each deposition was below 10^{-6} Pa. During deposition of the ZnO and AZO layers, highly purified argon and oxygen were used as process gas, which was inleted through a mass flow controller (MFC). Prior to the film deposition, pre-sputtering was performed for 10 min to remove the contaminants on the target surface. Al thin films with various thicknesses were deposited on the ZnO thin films using the RF sputter-deposition system. RF power was maintained at 150 W for AZO and 70 W for ZnO,

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respectively, for all the RF sputtering processes and the distance between the substrate and the target was fixed at 7 cm. The substrates were cleaned ultrasonically, first in acetone and then in methanol for 10 min prior to sputter deposition.

For the multilayer films prepared in such a way as described above, the AZO and ZnO film thicknesses were measured using a stylus profilometer (Dektak-3). On the other hand, the Al layer thickness was determined by atomic force microscopic (AFM) analysis (NS4A). The carrier concentration and the carrier mobility, and the resistivity of the TCO films were determined from Hall effect measurements (HEM-2000) using the van der Pauw geometry. The optical transmittance spectra of the films were obtained at room temperature in air using a dual beam ultraviolet/visible (UV/VIS) spectrometer (CARY 5E, VARIAN).

Prior to fabrication of multilayered structures, the optical, electrical, and morphological properties of ZnO, Al, and AZO films were investigated. Prior to deposition of Al and AZO films a thin ZnO layer (50 nm) was deposited on the glass, the role of which was to prevent degradation of the glass surface during the deposition of the Al/AZO thin films and to minimize the residual thermal stress, as well as to prevent diffusion or some kind of chemical reactions at the glass–film interface. In Fig. 1 the resistivity, the carrier concentration, and the carrier mobility of the AZO/Al/ZnO films have been plotted as functions of the Al interlayer thickness. As the Al interlayer thickness increases from 0 to 20 nm, the resistivity decreases all the way through the Al layer thickness range as the Al layer thickness increases. This is believed to be due to the presence of excess carriers (electrons) in the Al interlayer and not due to the carrier concentration in the AZO layer.

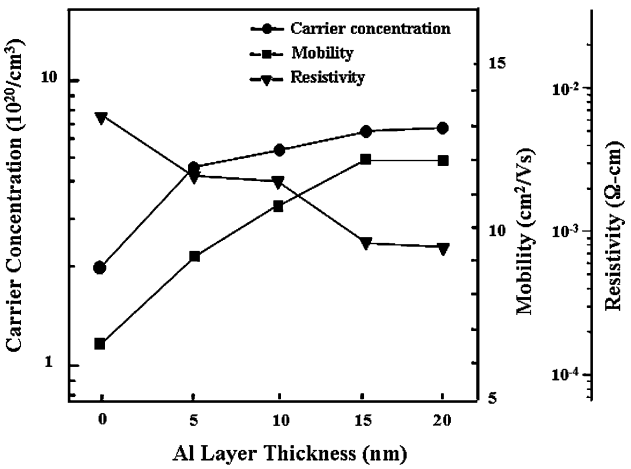

Fig. 1 The carrier concentration, the carrier mobility, and the electrical resistivity of the AZO/Al/ZnO multilayer film as a function of the Al interlayer thickness

The total sheet resistance R_s of an AZO/Al/ZnO multilayer structure tripled in parallel can be written as

$$\frac{1}{R_s} = \frac{t_{AZO}}{\rho_{AZO}} + \frac{t_{Al}}{\rho_{Al}} + \frac{t_{ZnO}}{\rho_{ZnO}}, \quad (1)$$

where ρ_{AZO}, ρ_{Al}, ρ_{ZnO}, t_{AZO}, t_{Al}, and t_{ZnO} represent the resistivities and the thicknesses of the AZO layer, the Al interlayer and the ZnO buffer layer, respectively. According to Eq. 1, if the Al interlayer thickness increases, the total sheet resistance will decrease. Since sheet resistance R_S is proportional to resistivity ($R_S = \rho/t$, where t is the thickness of a film) and the resistivity is inversely proportional to carrier concentration and carrier mobility $\mu(\rho = 1/q\ \mu n$, where q is carrier charge), we can rewrite Eq. 1 as

$$\mu_T = \frac{t_{AZO}\mu_{AZO}n_{AZO} + t_{Al}\mu_{Al}n_{Al} + t_{ZnO}\mu_{ZnO}n_{ZnO}}{t_T\, n_T}, \quad (2)$$

where the subscripts T, AZO, Al, and ZnO denote a total multilayer film, an AZO layer, an Al layer, and a ZnO layer, respectively. For the AZO/Al/ZnO/glass samples used in this work, μ_{Al} is far higher than μ_{GZO} and μ_{ZnO}. Thus, the carrier mobility of the multilayer film is strongly influenced by t_{Al}. Consequently, μ_T increases with t_{Al} up to a certain limit [17]. Figure 1 also shows that the variation trend of the carrier concentration of the film is similar to that of the carrier mobility. This trend is self-evident because the carrier concentration of the multilayer film can be expressed similarly to Eq. 2 as

$$n_T = \frac{t_{AZO}\mu_{AZO}n_{AZO} + t_{Al}\mu_{Al}n_{Al} + t_{ZnO}\mu_{ZnO}n_{ZnO}}{t_T\mu_T}. \quad (3)$$

We can confirm in Fig. 1 that both the carrier concentration and the carrier mobility, and thus the electrical resistivity of the AZO/Al/ZnO multilayer film strongly depend on the Al interlayer thickness. The resistivity decreases with Al layer thickness in the thickness range below 15 nm, although it nearly does not change with a further increase from 15 to 20 nm, suggesting that the optimum Al layer thickness is 15 nm.

Figure 2 shows a plot of the transmittance versus wavelength in the visible and near-infrared spectrum for the AZO/Al/ZnO films with various Al layer thicknesses. The Al layer mainly acts as a conductive layer, but its low transmittance is a drawback limiting the performances of the AZO/Al structure. The average transmittance in the visible range for all the films is 85%, more or less. In this figure we can see that the average visible transmittance of the AZO/Al film on ZnO-buffered glass does not change much with the Al layer thickness in the range from 0 to 15 nm. The high average transmittance (~85%) of this multilayer TCO film seems to be particularly attractive from a viewpoint of optical properties. A metal layer is

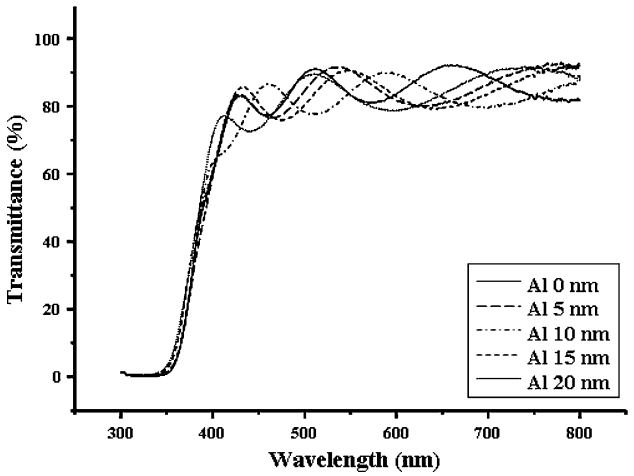

Fig. 2 The optical transmittance of AZO/Al/ZnO/glass samples with different Al interlayer thicknesses in a wavelength range from 300 to 800 nm

normally opaque. However, if a metal thin film is so thin that its thickness is less than a certain level, it becomes transparent. The transmittance of the film somewhat decreases as the Al layer thickness increases further up to 20 nm. Taking both the resistivity and the optical transmittance into consideration we may conclude that the optimum thickness of the Al underlayer for both the low resistivity and the high optical transmittance of the AZO/Al/ZnO multilayer film is 15 nm. Lastly, there is one thing to be mentioned here. As far as an Al layer is concerned, the problem of its reflectivity should be somehow solved. However, we can fortunately find a solution in the multilayer film structure itself consisting of AZO and Al layer. The AZO layer in the structure plays two important roles. Its relatively high conductivity assures a good electrical connection between two Al layers. On the other hand, this transparent layer may act as antireflective coatings for a certain Al layer thickness, reducing the high reflectivity of the Al layer [15].

In conclusion, the electrical properties of AZO films can be significantly improved by using an ultrathin Al interlayer. The optimum Al interlayer thickness with which a low resistivity can be obtained without nearly harming the optical transmittance in the visible light region is found to be 15 nm. The resistivity and the transmittance of the AZO(200 nm)/Al(15 nm)/ZnO(50 nm) deposited on glass by sputtering are $8.1 \times 10^{-4}\ \Omega$ cm and $\sim 83 \pm 7\%$, respectively.

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References

1. Pei ZL, Sun C, Tan MH, Xiao JQ, Guan DH, Huang RF, Wen LS (2001) J Appl Phys 90:3432
2. Tabuchi K, Wenas WW, Yamada A, Kakahashi K (1993) Jpn J Appl Phys 1:3764
3. Lamp U, Muller J (1989) Sens Actuators 18:269
4. Horsthuis WHG (1986) Thin Solid Films 137:185
5. Hong JS, Rhee BR, Kim HM, Je KC, Kang YJ, Ahn JS (2004) Thin Solid Films 467:158
6. Lewis BG, Paine DC (2000) MRS Bull 25:22
7. Elvira F, Alexandra G, Vitor A, Antonio M, Hugo A, Luıs P, Isabel F, Rodrigo M (2003) Thin Solid Films 442:121
8. Assuncao V, Fortunato E, Marques A, Aguas H, Ferreira I, Costa MEV, Martins R (2003) Thin Solid Films 427:401
9. Yim K, Lee C (2006) Cryst Res Technol 41:1998
10. Yim K, Kim HW, Lee C (2007) Mater Sci Technol 23:108
11. Kim S, Lee W, Lee C (2007) Mater Sci Technol 23:303
12. Kim H, Horwitz JS, Qadri SB, Chrisey DB (2002) Thin Solid Films 107:420
13. Sahu DR, Huang JL (2006) Thin Solid Films 515:876
14. Hu YM, Lin CW, Huang JCA (2006) Thin Solid Films 497:130
15. Bertran E, Corbella C, Vives M, Pinyol A, Person C, Porqueras I (2003) Solid State Ionics 165:139
16. Sahu DR, Huang JL (2006) Thin Solid Films 515:876
17. Tokumoto MS, Smith A, Santilli CV, Pulcinelli SH, Craievich AF, Elkaim E, Traverse A, Briois V (2002) Thin Solid Films 416:284