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Effect of Thermal Annealing on the Electrical Properties of In-Si-O/Ag/In-Si-O Multilayer

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Transparent conductive multilayers have been fabricated using transparent amorphous Si doped indium oxide (ISO) semiconductors and metallic Ag of ISO/Ag/ISO. The resistivity of a multilayer is dependent on the middle layer thickness of silver. The thickness of the Ag layer is fixed at 11 nm and takes into account cost and optical transmittance. As-deposited ISO/Ag (11 nm)/ISO multilayer shows a measured resistivity of 7.585×10^{-5} Ω cm. After a post annealing treatment of 400℃, the resistivity is reduced to 4.332×10^{-5} Ω cm. The reduction of resistivity should be explained that the mobility of the multilayer increased due to the optimized crystalline, meanwhile, the Hall concentration of the multilayer showed an obscure change because the carriers mainly come from the insert of the Ag layer.

Keywords: Si doped InO, Thermal annealing, Electrical property

1. INTRODUCTION

Tin doped indium oxide (ITO) is a commercial transparent conductive electrode material used in many applications because of its high transmittance in visible regions and high electrical conduction properties. However, ITO thin films show polycrystalline structures deposited at room temperature (RT), causing their fragile mechanical characteristic [1]. To improve the stability of In oxide based electrode, a multi-functional doping agent of Si was introduced to substitute for Sn dopant: first, same with Sn, Si substitute into the In3+ site as n-type dopant [2], then, it is reported that Si doping plays a role of carrier suppressor to improve the electrical stability of thin films by controlling the oxygen defects [3], at the same time, Si doping can prevent the phase transition of In_2O_3 from amorphous to crystalline, then improve the mechanical strength of In oxide based thin films [2].

Recently, oxide-metal-oxide (OMO) is widely studied as transparent electrodes in solar cells, owing to its excellent transparency in visible regions and metal-like electrical conductivity. Due to the addition of the middle metal layer, both the electrical and optical properties are optimized. On electrical aspects, a parallel circuit model of R_{oxide} // R_{metal} // R_{oxide} is often used to explain the resistance models of OMO structures [4]. Due to the resistances model, the metal layer is the main path of electrical conduction.

2. EXPERIMENTS

Amorphous ISO films used as bottom/top oxide layers were prepared on glass (Corning 1737) substrate by DC magnetron sputtering method at room temperature. The sputtering conditions for a-ISO layers, such as base pressure, working pressure, Ar gas flow rate, sputtering rate, and sputtering power were 5.0×10^{-6} Torr, 4.0×10^{-3} Torr, 30 sccm, 9.7 Å/s and 30 W, respectively. Deposition of a-ISO films was conducted using a ceramic ISO target. The middle Ag film sandwiched between two outer oxide layers has also grown due to the DC method at room temperature. Thickness of each top and bottom a-ISO layer in ISO/Ag/ISO (OMO) structure was about 40 nm. The thickness of Ag layers has been systematically changed, ranging from 5 nm to 15 nm. The structural property of OMO films has been characterized by using X-ray diffraction (XRD), atomic force microscopy (AFM).

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The visible and IR transmittance was analyzed by UV?VIS optical spectrometer (Cary 5000 UV-Vis-NIR, Agilent).

3. RESULTS AND DISCUSSION

Figure 1 shows the XRD results obtained from an ISO/Glass substrate as a function of annealing temperature, XRD plots of as-deposit ISO films indicating a completely amorphous structure. As the temperature increases, two sets of diffraction peaks are strengthened, which are (222), (400), (440) from bixbyite In_2O_3 and (202) from SiO_2. It is reported that in ISO thin film, Si^{4+} substitutes into the In^{3+} site as n-type dopant [2]. From the XRD results, we can see that the temperature increase led to a SiO_2(202) peak strengthen, indicating doped Si^{4+} got away from the In_2O_3 structure and combined with oxide.

Carrier concentration in ISO single layer films showed a sharp decrease of two orders of magnitude from $4.34\times10^{20}/cm^3$ to $3.3\times10^{18}/cm^3$ measured by Hall method as shown in Fig. 2 depending on annealing temperature. Increase of annealing temperature results in the increase of resistivity which is mainly caused by decreased carrier concentration because oxygen defects should be reduced as a result of increased indium-oxide binding [5]. The doped Si^{4+} would get away from the In_2O_3 structure then combined with oxide, leading to the decrease in carrier concentration caused by the decrease of doped Si^{4+}. However, mobility has increased, which can be explained by the degenerated semiconductor properties of as-deposited ISO film, in where mobility was limited by electron-phonon scattering effect [6]. In this work, mobility increased observably from 18.9 $cm^2/(Vs)$ to 63.3 $cm^2/(Vs)$ owing to the lower probability of scattering caused by the decrease of free carriers.

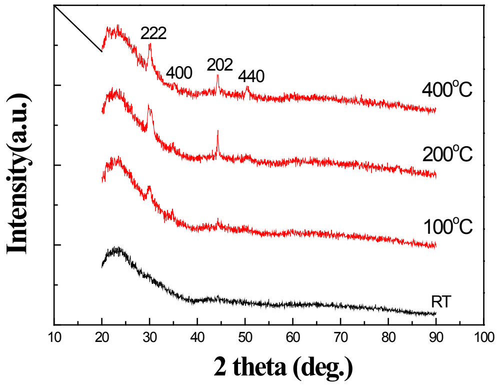

Fig. 1. X-ray diffraction (ω - 2 θ) patterns for as-deposit ISO film (black line), and 100℃, 200℃, 400℃ annealed ISO film(red line).

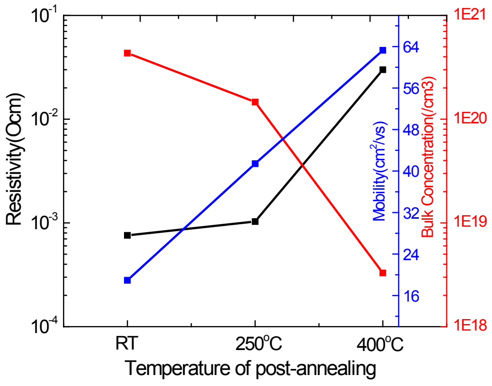

Fig. 2. Measured resistivity, mobility, and Hall concentration of the ISO films as a function of annealing temperature.

To explain the electrical properties of these tri-layers, a parallel circuit model of R_{oxide} // R_{metal} // R_{oxide} was modeled, as shown in Fig. 3. Based on this model, the resistivity of OMO can be defined as $1/R_{OMO}= 1/R_{Oxide}+1/R_{Metal}+1/R_{Oxide}$.

From Fig. 2, the resistivity of single ISO layer showed a 7.585×10-4 Ω cm measured at room temperature. For the comparison, the single metallic silver layer has a much lower resistance value. Based on the parallel circuit formula, it indicates that the metal layer is the main path of electrical conduction in the structure of OMO, due to the huge difference of resistance.

Figure 4 shows the electrical properties of as-deposited ISO/Ag/ISO tri-layer thin films measured by four point probe method. In this method, the resistivity of the thin film is valued by two parameters, the bulk concentration and the free carrier mobility as follows:

$$\rho=1/ne\mu \tag{1}$$

where ρ is the resistivity, n is the number of charge carriers, e is the charge of the carrier, and μ is the carrier mobility.

As the thickness of Ag layer's increase, both the carrier concentration and the mobility increased, which proved that metallic Ag layer is the main conduction path. And it is important to note that the curve of bulk concentration almost increases linearly as increasing the thickness of Ag layer.

The electrical properties of ISO/Ag (11 nm)/ISO thin films have been measured as shown in Fig. 5. The Hall concentration shows obscure change from maximum $7.169\times10^{21}/cm^3$(at RT) to $6.551\times10^{21}/cm^3$(at 250℃) unlike the sensitive changes of ISO single layer as shown in Fig. 2. This phenomenon can be explained

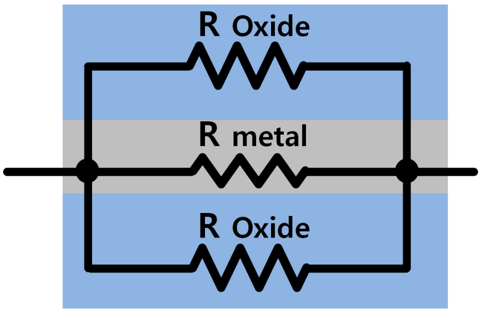

Fig. 3. A parallel circuit model of oxide/metal/oxide structure.

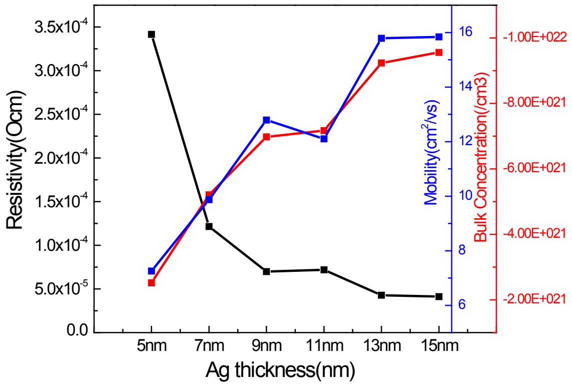

Fig. 4. Hall measurement result of ISO/Ag/ISO thin films as a function of Ag thickness.

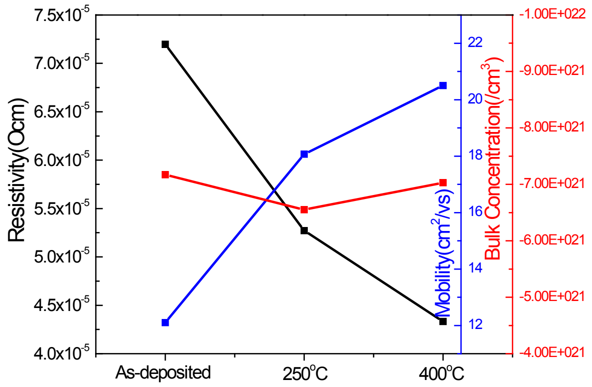

Fig. 5. Hall measurement result of ISO/Ag(11 nm)/ISO thin films as a function of post-annealing temperature.

that the measured bulk concentration is mainly from the middle silver-layer, due to the huge difference of carrier concentration in silver layer (above 10^{22}/cm^3) and ISO layer (measured as 3.3×10^{18}/cm^3 to 4.3×10^{20}/cm^3 as shown in Fig. 2) [7]. Under such conditions, Hall concentration could be fixed by adjusting the thickness of the Ag layer, meanwhile, mobility can be optimized as increasing post-annealing temperature as shown in Fig. 5, and it is reported that as post annealing temperature increased, Ag films becomes crystallized, thanks to grain growth in Ag layers the grain boundary's carriers scattering decreased and consequently the mobility increased [8], As a conclusion, post-annealing is a simple and effective method to control the electrical properties of the ISO/Ag/ISO multilayer.

4. CONCLUSIONS

Ag layer played an important role to improve the electrical properties of ISO/Ag/ISO tri-layer, and we manufactured as-deposited ISO/Ag(11 nm)/ISO which shows a resistivity value of 7.198×10^{-5} Ωcm by adjusting the thickness of Ag. By post-annealing treatment in air, the resistivity of ISO/Ag(11nm)/ISO showed 4.332×10^{-5} Ωcm, and commercial indium tin oxide with thickness of about 150nm shows electrical resistivity of 6-7× 10^{-5} Ωcm [9], the resistivity of ISO/Ag/ISO should be the applicable level for transparent conductive electrode.

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