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ZnO/Ag/ZnO multilayer films for the application of a very low resistance transparent electrode

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Abstract

Transparent conductive ZnO/Ag/ZnO multilayer electrodes having much lower electrical resistance than the widely used transparent electrodes were prepared by simultaneous RF magnetron sputtering of ZnO and DC magnetron sputtering of Ag. An Ag film with different thickness was used as intermediate metallic layers. The optimum thickness of Ag thin films was determined to be 6 nm for high optical transmittance and good electrical conductivity. With about 20–25 nm thick ZnO films, the multilayer showed high optical transmittance in the visible range of the spectrum and had color neutrality. The electrical and optical properties of the multilayers were changed mainly by Ag film properties. A high quality transparent electrode, having sheet resistance as low as 3 ohm/sq and high transmittance of 90% at 580 nm, was obtained and could be reproduced by controlling the preparation parameter properly. The above property is suitable as transparent electrode for dye sensitized solar cells (DSSC).

Keywords: ZnO; Transparent electrode; Multilayer

1. Introduction

Transparent conducting oxides (TCOs) such as impurity doped indium oxides, tin oxides, zinc oxide systems have been used in numerous optoelectronic devices such as flat panel displays [1,2] and photo voltaic solar cells [3,4]. However, their resistivity is rather high in some cases to adapt as a transparent electrode for improved practical application. Method of depositing thin film with reduced resistivity is being investigated in order to accommodate the increasing technological demand for large area devices with improved performance. Recently, for the improvement of the conductivity of transparent electrode, ITO-metal–ITO multilayer systems are used. A thin metal layer of about 10 nm thickness was embedded between two ITO layers [5–7]. These IMI structures

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have very low sheet resistance, high optical transparency in the visible range, relatively lower thickness than single-layered TCO film and better durability than single-layered metal film [8–11]. However, the major cost factors, in the production of TCO are the extremely high target cost of ITO [12]. One of the most potential candidates to substitute ITO film is being the ZnO due to its non-toxicity [13], low cost [14], material abundance [15], high stability against hydrogen plasma and heat cycling [16]. The structural characteristics, electrical and optical properties of the ZnO films have been investigated widely [17–21] while ZnO-based multilayers are still under investigation.

It is well known that the optical and electrical properties of very thin metal films depend considerably on their structures. To get bulk like properties, the metal film should form a continuous structure, although they must be thin for high transmittance. Ag metal films, which have highest conductivity of all metals has been already used for ITO-based [22,5–7] multilayer for lower resistance good transparent conducting electrode. However, there is no report related to Ag and ZnO-based multilayer for the application of low resistance transparent electrode. Therefore, we used silver and developed ZnO/Ag/ZnO (ZAZ) transparent conductive film. The influence of the preparation process on the properties of the film was investigated.

2. Experimental

The thin films of ZnO and ZnO/Ag/ZnO structures were sputter deposited on glass (corning 1737F) using a zinc oxide (99.9995 purity, 7.62 cm diameter, 0.64 cm thickness, target materials Inc.) and metal Ag targets (99.999% purity, 7.62 cm diameter, 0.64 cm thickness, target materials Inc.) in an inline magnetron sputter deposition system equipped with DC and RF power suppliers. The glass substrate was ultrasonically cleaned in acetone, rinsed in deionized water and subsequently dried in flowing nitrogen gas before deposition. The sputtering was performed in argon atmosphere with a target to substrate distance of 53 mm. The sputtering was carried out at a pressure 6×10^{-3} Torr in pure Ar with varying sputtering parameters such as argon flow rate and RF/DC power. The rotating speed of the substrate was 18 rpm. The thickness of the ZnO layer was varied between 20 and 100 nm and those of Ag were between 1 and 15 nm. Film thickness was measured using a surface profiler (Alpha-step 500, TENCOR) and FE-SEM (XL-40 FEG field emission scanning electron microscope). Conventional θ–2θ XRD studies on the films were carried out in Regaku (D/MAX 2500) diffractometer using Cu Kα radiation to investigate the crystallinity and crystal orientation of the films. Sheet resistance was measured using 4-point probe method. Optical transmittance was measured in the range of 300–800 nm by UV–vis–IR spectrophotometer (Hewlett Packard 8452A diode array spectrophotometer).

3. Results and discussion

The crystalline structure of different multilayers was determined by XRD measurements. Fig. 1 presents the XRD patterns of as deposited ZnO and ZAZ multilayers. A strong (0 0 2) peak along with (1 0 3) was seen for ZnO film. Strong (0 0 2) preferential orientation, indicating polycrystalline nature of the film. In case of ZAZ multilayer, another (1 0 2) peak was developed but ZnO grains are mainly (0 0 2) aligned corresponding to wurtzite structure of ZnO [23]. Silver had (1 1 1) orientation. However, with increase of thickness of Ag layer, additional

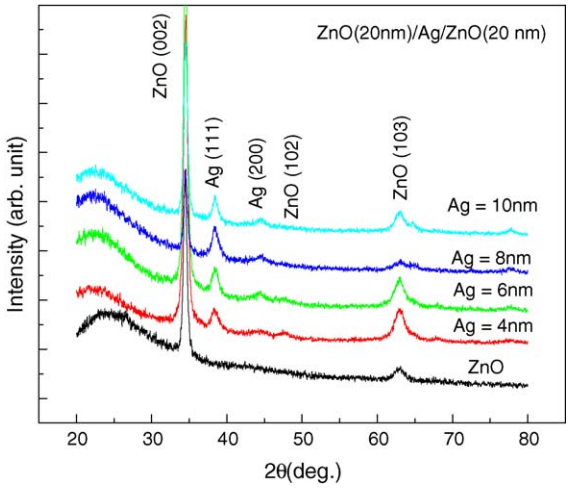

Fig. 1. The XRD patterns of as deposited ZnO and ZnO/Ag/ZnO multilayers.