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Study of Cu-based Al-doped ZnO multilayer thin films with different annealing conditions

Chien-Hsun Chu[a], Hung-Wei Wu[b,*], Jow-Lay Huang[a,c,d]

[a]Department of Materials Science and Engineering, National Cheng Kung University, Tainan 70101, Taiwan
[b]Department of Computer and Communication, Kun Shan University, Tainan 71003, Taiwan
[c]Department of Chemical and Materials Engineering, National University of Kaohsiung, Kaohsiung 81148, Taiwan
[d]Research Center for Energy Technology and Strategy, National Cheng Kung University, Tainan 70101, Taiwan

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Abstract

AZO/Cu/AZO multilayer thin films produced under different annealing conditions are studied in this paper, to examine the effects of atmosphere and annealing temperature on their optical and electrical properties. The multilayer thin films are prepared by simultaneous RF magnetron sputtering (for AZO) and DC magnetron sputtering (for Cu). The thin films were annealed in a vacuum or an atmosphere of oxygen at temperatures ranging from 100 to 400 °C in steps of 100 °C for 3 min. High-quality multilayer films (at Cu layer thickness of 15 nm) with resistivity of 1.99×10^{-5} Ω-cm and maximum optical transmittance of 76.23% were obtained at 400 °C annealing temperature in a vacuum. These results show the films to be good candidates for use as high quality electrodes in various displays applications.

Keywords: Multilayer; AZO/Cu/AZO; Rf sputter; Thin films; Annealing conditions

1. Introduction

In recent years AZO thin films have attracted much attention as transparent conductive oxide (TCO) materials. Many studies have shown that such films exhibit high conductivity and transmittance in the visible region, and high reflectivity in the infrared region [1,2]. However, the inherent limitation of such films with regard to increasing the conductivity has led to the development of multilayer structures. Due to the growing demand for high performance TCO films, it is thus necessary to investigate how superior electrical and optical properties of such films can be obtained.

AZO/metal/AZO multilayer films with high optical and electrical properties have been reported in the literature [3–5], and these structures show very lower resistivity and a selective high transparent effect by suppressing reflection from the metal layer. In [6], the multilayer films were deposited by DC magnetron sputtering at room temperature. The multilayered thin film with a Cu layer thickness of 7 nm displayed the highest figure of merit of 4.82×10^{-3} Ω^{-1}, with a low sheet resistance of 21.7 Ω/sq and an acceptable visible transmittance of 80%. These works has provided many good results for high quality thin film development and inspiring us to further study this topic. Simultaneous optimization of conductivity and transparency presents a challenge in multilayer film deposition. In this study, we use copper (Cu) films as an intermediate metal layer, as the Cu block has very low resistivity of about 1.67×10^{-6} Ω-cm [7]. The most important role of the top and bottom dielectric layers is to suppress the light reflection of the metal layer, and further to achieve the maximum light transmittance [8]. The Cu layer can improve the electrical properties of multilayer films, and some studies have shown the properties of tri-layer structures with Cu [9,10]. Annealing is an effective method to improve the electrical and optical properties of thin films, which change at different atmospheres and temperatures [11,12]. Moreover,

*Corresponding author. Tel.: +886 6 2051237; fax: +886 6 2050009.
E-mail addresses: hwwu@mail.ksu.edu.tw, qqq25q@gmail.com (H.-W. Wu).

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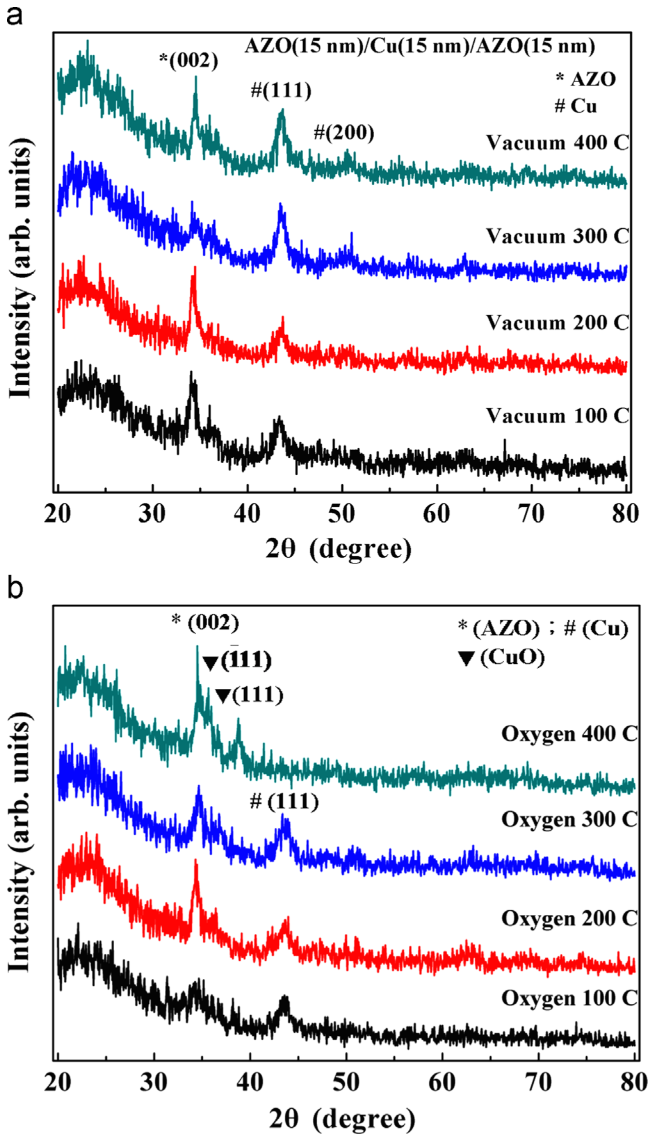

Fig. 1. XRD plots of the AZO/Cu/AZO films with (a) vacuum and (b) oxygen atmosphere annealing.

while previous studies have reported the properties of AZO/Cu/AZO films, there is still a lack of knowledge with regard to copper's impact on their structural, optical and electrical properties.

The goal of this study is thus to investigate the optical and electrical properties of AZO/Cu/AZO films produced under different annealing conditions. The multilayer thin films are prepared by simultaneous RF magnetron sputtering (for AZO) and DC magnetron sputtering (for Cu). The sputtering process is considered to be the most favorable deposition method, the high deposition rate can produce highly uniform films [13]. High-quality multilayer films (at Cu layer thickness of 15 nm) with a resistivity of 1.99×10^{-5} Ω-cm and maximum optical transmittance of 76.23% are obtained in this study at a 400 °C annealing temperature and in vacuum. These results show the films to be good candidates for use as high quality electrodes in various displays applications.

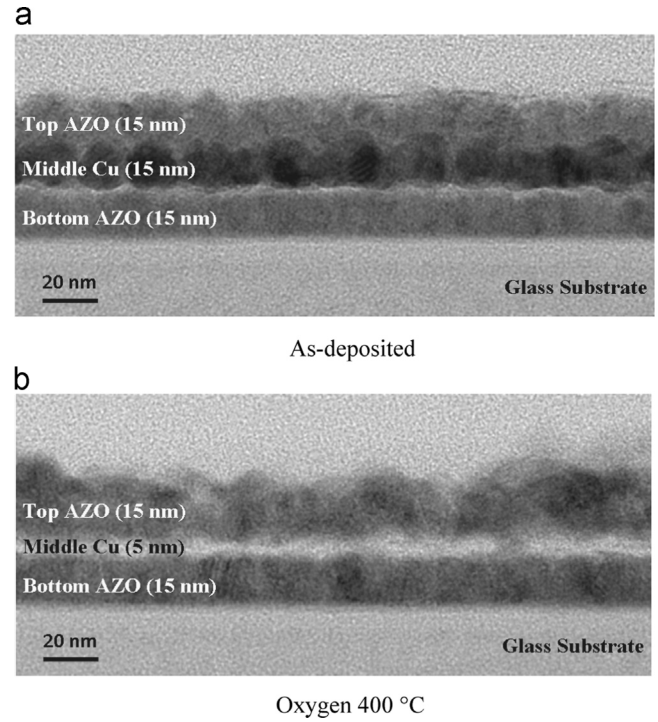

Fig. 2. Cross-sectional high resolution transmission electron microscope (HR-TEM) images for the as-deposited films and at an annealing temperature of 400 °C in an oxygen atmosphere.

2. Experimental procedures

AZO/Cu/AZO tri-layer films were deposited on the glass substrate (Corning Eagle XG; $20 \times 20 \times 0.5$ mm^3) by rf magnetron sputtering using an AZO ceramic target (99.9995% purity, 200 mm diameter, 50 mm thickness Al_2O_3:ZnO = 2:98 wt%) and dc magnetron sputtering using a Cu metal target (99.999% purity, 7.62 cm diameter, 0.64 cm thickness). The glass substrates were ultrasonically cleaned in an ethanol/acetone solution and then rinsed in deionized water. AZO thin film sputtering was performed in an argon atmosphere (purity: 99.99%) with a target-to-substrate distance of 15 cm. Initially, the rf sputtering chamber was evacuated to a base pressure of 4×10^{-4} Pa with a turbo molecular pump. The rf power and the substrate temperature were kept at 250 W and 70 °C, respectively, while there was an Ar flow ratio of 180 sccm and working pressure of 2.4 Pa. The dc sputtering chamber was evacuated to a base pressure of 1.33×10^{-4} Pa with a turbo molecular pump. The dc power and substrate temperature were kept at 100 W and 200 °C, respectively, and there was an Ar flow ratio of 12 sccm and working pressure of 0.4 Pa. The deposition rates of the AZO and Cu layers were 0.028 nm/s and 0.556 nm/s, respectively. The thickness of the AZO layer was 15 nm, while that of the Cu was 15 nm. The AZO/Cu/AZO films were annealed via rapid thermal annealing (RTA) and processing in vacuum or an oxygen atmosphere. The annealing temperatures of the films were set at 100, 200, 300 and 400 °C for 3 min. The heating rate was controlled to 10 °C/min and the gas flow rate was maintained at 1000 sccm in 4 Pa for all experiments. Conventional θ–2θ XRD studies of