

## Dependence of ZnO thickness on the electrical and optical properties of ZnO-Cu-ZnO multilayers for optoelectronic application

Hiba S. Rasheed<sup>1,2,\*</sup>, Naser M. Ahmed<sup>1</sup>, M. Z. Matjafri<sup>1</sup>, Fayroz A. Sabah<sup>1,3</sup>

<sup>1</sup>*Institute of Nano-Optoelectronics Research and Technology (INOR), School of Physics, Universiti Sains Malaysia, 11800 Penang, Malaysia*

<sup>2</sup>*Department of Physics, College Of Education, Al-Mustansiriya University, Baghdad, Iraq*

<sup>3</sup>*Department of Electrical Engineering, College Of Engineering, Al-Mustansiriya, Baghdad, Iraq*

**Abstract:** in this work ZnO/Cu/ZnO transparent conductive multilayer films are prepared ZnO using RF sputtering and Cu film using DC sputtering. The properties of the stacks structure are studied at different thickness of ZnO. The results show that the sheet resistance of the film stacks decreases with an increase of ZnO thickness and increased further with increase Fig. of merit. However, the transmittance of the structure increased with the decrease of ZnO thickness. Good transparent conductive film of sheet resistance was 84.29 ohms/sq while the transmittance of 53% was found at a 30/5/30. The performance of the multilayer film was evaluated using a Fig. of merit. The properties of these film stocks good for different applications in optoelectronic devices.

**Key words:** RF sputtering; Cu target; N-type; TCO; Multilayer

### 1. Introduction

Transparent conductive oxide (TCO) films have been extensively studied because of their technological applications in solar cells, optoelectronics, gas sensors, flat panel displays and thin film resistors (Sahu and Huang, 2006). The efficiency and performance of these devices depend on the electrical and optical properties of the TCOs. The most widely used TCOs are impurity-doped indium oxides, tin oxides and zinc oxides which offer a commercially acceptable performance in terms of conductivity, transmittance, environmental stability, reproducibility and surface morphology (Paul et al., 2003; Leng et al., 2010). However, their resistivity is rather high in some cases to adapt as a transparent electrode for improved practical application. It is therefore necessary to look for a transparent conductive film having lower resistance and higher transmittance. Recently, a combination of semiconducting, dielectric materials and metal multilayers is used to achieve high transparent conducting oxides (Lou et al., 2009). Metal-based indium tin oxide (ITO) and ZnO multilayer structures are reported as good transparent conducting electrodes (Raniero et al., 2006; Park et al., 2009). Due to the high target cost of ITO (Wang and Diao, 2010), ZnO-based multilayers are the potential material for large scale production of good TCO films have been extensively studied because the potential material for large scale production of good TCO. However, simultaneous optimization of conductivity and transparency presents a considerable challenge

in ZnO-based multilayer film deposition. It is also well known that films of the same material deposited under different conditions possess different electrical and optical properties (Dose et al., 1984; Cho et al., 2006). In order to do further improvement in the conductivity, the ZnO-based single layer Metal thin film for the metal types, there are many excellent conductivity met, Cu, Al, Fe and so on. Metal Cu with It has excellent conductivity, and the price is relatively compare with Ag has a distinct advantage the electrical properties of the structure as you see in the figure very high dependent on the mobility and concentration of the flow of the electrons and the electrical characteristic will change by increasing the thickness of Cu or the ZnO, and this depend on the work function of the both material.

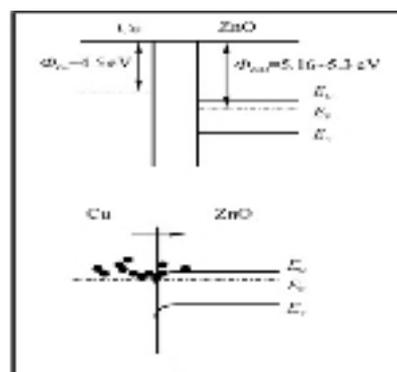


Fig. 1: Schematic diagram of band gap energy of ZnO and Cu.

\* Corresponding Author.

In this study, Cu metal layer is sandwiched layer between the ZnO films by sputtering method at room temperature to prepare a ZnO / Cu / ZnO transparent conductive multilayers film, designed and obtain the optimizing Cu layer thickness strip at room temperature to get high performance photovoltaic element under transparent conductive film.

**2. Experimental**

Thin films of ZnO/Cu/ZnO were sputter deposited on glass microscopic slide using a zinc oxide (99.9995% purity, 7.62 cm diameter, 0.64 cm thickness, target materials) and metal Cu targets (99.999% purity, 7.62 cm diameter, 0.64 cm thickness, target material) in an in-line magnetron sputter deposition system equipped with DC and RF power (55W ). 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 at a pressure of  $6 \times 10^{-3}$  Torr in pure Argon atmosphere with a target-to-substrate distance of 53mm. A Cryo pump, coupled with a rotary pump used to achieve  $1 \times 10^{-6}$  Torr pressure before introducing argon gas (99.9995%, Lien Hwa gas co.). The rotating speed of the substrate was 18 rpm. The

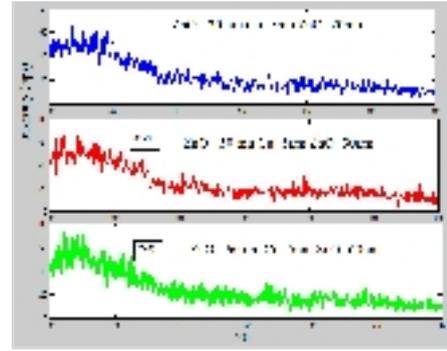
The thickness of the ZnO layer was 60nm and that of Cu was 5 nm taken for this study due to its superior performance at room temperature [18]. Film thickness was measured using FE-SEM (Nova nanosem450 field emission scanning electron microscope) Thickness of the ZnO and Cu layers was confirmed through deposition rate Surface roughness were determined and surface images were taken by Atomic Force Microscope (AFM, Sheet resistance was measured using(Accent optical technologies Inc HL 5500pc-hall effect four-point probe method). Optical transmittance was measured in the range of 200–800 nm by UV–VIS–NIR spectrophotometer.(Agilent Technologies Cary series UV-VIS-NIR spectrophotometer.

**3. Results and discussion**

Fig. 2 presents the XRD pattern of the multilayer grown with fixed thickness of Cu layer and with different thickness of ZnO layer. With increase of the thickness of ZnO layer, intense

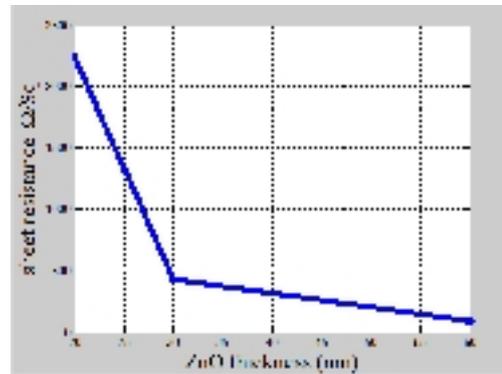
(0 0 2) orientation peaks are observed. The intensity of (1 0 3) peak decreases and (0 0 2) increases when thickness of ZnO layer changed from 20 to 50 nm. Crystallinity increased with increase of thickness of ZnO layer. The observed diffraction pattern indicates the polycrystalline structure but with preferential orientation along c-axis. However, other peaks appear for further increase of thickness of ZnO layer and the location of measured diffraction peak shows a small shift towards the lower diffraction angle. So shift of (0 0 2) peaks towards lower diffraction angle with increasing thickness of ZnO layer indicates that there is distortion of

crystallites (Cho et al., 2006; Zhu et al., 2006). So better crystallinity with preferred c-axis orientation was observed for 50 nm ZnO layer.

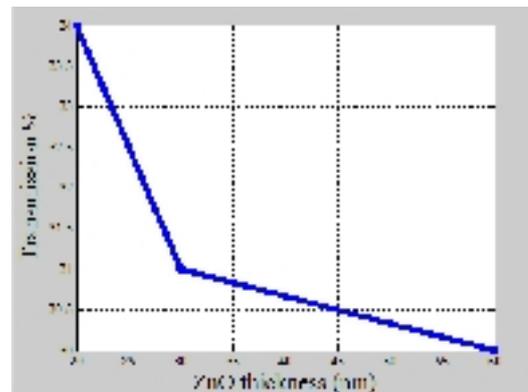


**Fig. 2:** XRD pattern of ZnO/Cu/ZnO multilayer with fixed thickness of Cu layer (5 nm) and with different thickness of ZnO layer.

Fig. 2 Refer that the sheet resistance of ZnO–Cu–ZnO multilayers depending the thickness of ZnO layer. The thickness of the inter layer films was set to be constant at 5 nm and the thickness of ZnO layer was varied from 20 to 60 nm. The sheet resistance of the multilayer films was lower than the resistance of single ZnO layer (film thickness 60 nm) which lacks copper layer. Furthermore, the multilayer film resistance decreases as the thickness of ZnO layer increases. With the insertion of a 5 nm copper layer between the ZnO layers, a reduction in resistance by approximately 95% relative to that of single layer ZnO film of the same thickness was achieved.



**Fig. 3:** Dependence of the sheet resistance of ZnO/Cu(5nm)/ZnO multilayer film on the thickness of the ZnO layer.



**Fig. 4:** Depended transmission of the multilayer on the thickness of ZnO.

The optical properties of ZnO are also very important for the performance of the stack. ZnO causes an increase in the transmission with decrease in the thickness.

The relationship between hall mobility and ZnO thickness was investigated in Fig. 5, it refers that when ZnO thickness =20nm the hall mobility is maximum value =27.5(cm<sup>2</sup>/v.s).

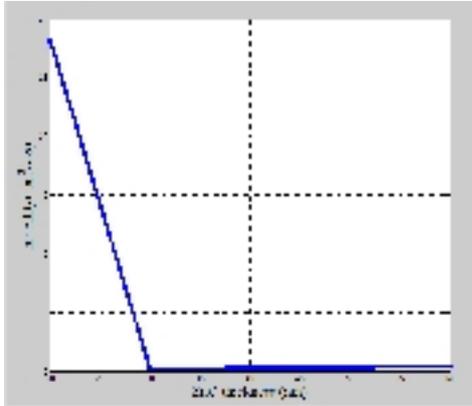


Fig. 4: Relationship between hall mobility and ZnO thickness.

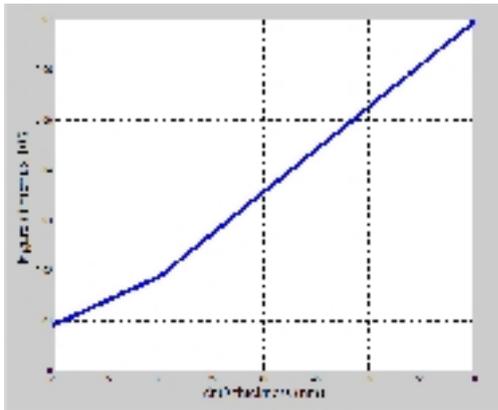


Fig. 5: The Fig. of merit for the ZnO/Cu (5 nm)/ZnO multilayer films with different ZnO layer thicknesses.

In order to value the TCO films and compare their photoelectric performance, the Fig. of merit (FOM) is applied.  $FOM = T^{10}/RS$  was defined by Haacke (Zhu et al., 2006), where T is the optical transmittance and Rs is the sheet resistance. Fig. 5 shows the Fig. of merit for the multilayer films with ZnO thickness, and it indicated that the multilayer had a high Fig. of merit with increasing ZnO thickness.

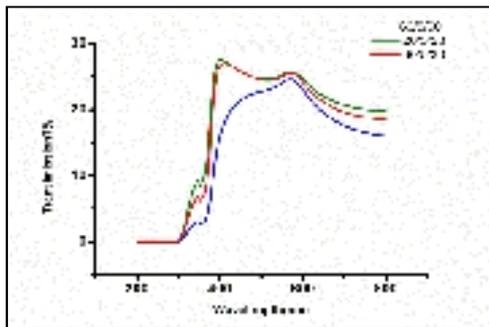


Fig. 6: Dependence of transmittance of the ZnO/Cu (5 nm)/ZnO multilayer films on ZnO layer thickness.

Fig. 6 presents the optical transmittance spectra of the ZnO/Cu/ZnO multilayer films with different ZnO layer thicknesses for fixed thickness of Cu layer (5 nm). It is seen that as the thickness of ZnO (top and bottom) layers increases, the transmittance decreases in the UV region and the absorption edge is shifted to longer wavelength regions. When the thickness of the ZnO layers changes in the range of 300-600 nm, the multilayer films represent a better optical property; their transmittance spectra have a relatively flat plateau in the entire visible region.

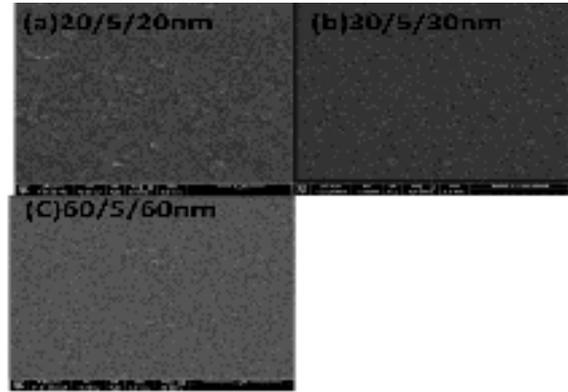


Fig. 7: FESEM micrographs (ZnO/ Cu(5nm) /ZnO) films deposited with various deposition Thickness of ZnO.

Fig. 7 shows FESEM images of the three multilayers; and it refer to the morphology of ZnO surface that is nanostructure during the first stage of film formation. The multilayer of thickness 60-5-60nm appears an uniformly distribution on the surface, which indicate that the crystallinity of the top layer ZnO is better than before.

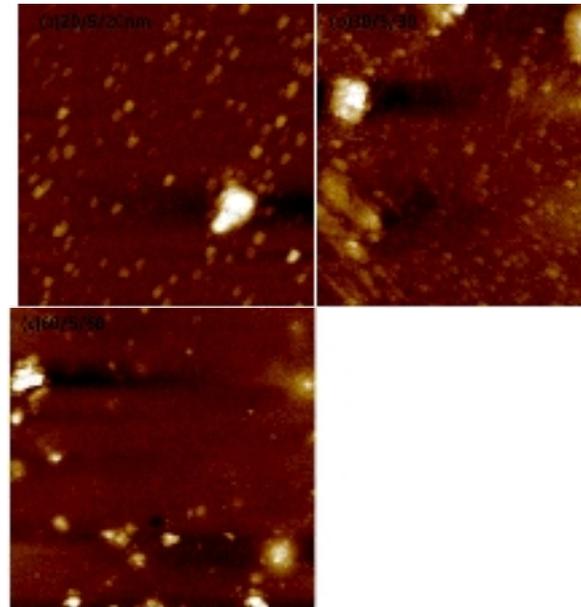


Fig. 8: AFM images of the multilayer films.

Fig. 8 shows the AFM images of multilayer films deposited with different thickness of ZnO and fix thickness of Cu., roughness of multilayer films decreases with increase of ZnO thickness.

#### 4. Conclusion

Different layer stacks of ZnO–Cu–ZnO have been examined. The influence of metal film thickness on optical and electrical properties was investigated. It was found that the layer stack can be optimized to have sheet resistance below 10  $\Omega$ /sq. at a total light transmission over 25% at 400 nm. Under these conditions, the electrical properties of ZnO are of minor importance. The quality of the multilayer film is determined by the properties of the Cu film instead of ZnO properties.

### Acknowledgment

We gratefully acknowledge the support of University Sains Malaysia and RU grant number; RKGS 203/PFIZIK/6711353.

### References

- D.R. Sahu, J.L. Huang, Thin Solid Films 515 (2006) 876.
- D.Y. Zhang, P.P. Wang, R.I. Murakami, X.P. Song, Appl. Phys. Lett. 96 (2010) 233114.
- G. Haacke, J. Appl. Phys. 47 (1976) 4086.
- G.K. Paul, S. Bandyopadhyay, S.K. Sen, S. Sen, Mater. Chem. Phys. 79 (2003) 71.
- H.Y. Liu, V. Avrutin, N. Izyumskaya, Ü. Özgür, H. Morkoç, Superlattices Microstruct. 48 (2010) 458.
- J. Leng, Z.N. Yu, W. Xue, T. Zhang, Y.R. Jiang, J. Zhang, D.P. Zhang, J. Appl. Phys. 108(2010) 073109.
- J. Lou, M. Bao, B.J. Ye, H.M. Weng, H.J. Du, Z.B. Zhang, J. Opt. A: Pure Appl. Opt. 11 (2009) 085501.
- L. Raniero, I. Ferreira, A. Pimentel, A. Gonçalves, P. Canhola, E. Fortunato, R.
- M. Born, E. Wolf, Principles of OpticsSixth Edition, , 1980, p. 55.
- Martins, Thin Solid Films 511 (2006) 295.
- P. Yianoulis, M. Giannouli, J. Nano. Res. 2 (2008) 49.
- P. Zhao, W.T. Su, R. Wang, X.F. Xu, F.S. Zhang, Physica E 41 (2009) 387.
- S.H.K. Park, C.S. Hwang, M. Ryu, S. Yang, C. Byun, J. Shin, J.I. Lee, K. Lee, M.S. OhS. Im, Adv. Mater. 21 (2009) 678.
- T. Wang, X.G. Diao, P. Ding, Appl. Surf. Sci. 257 (2010) 3748.
- V. Dose, W. Altmann, A. Goldmann, U. Kolac, J. Rogozik, Phys. Rev. Lett. 52 (1984) 1919.
- X. Wu, T.J. Coutts, W.P. Mulligan, Vac. Sci. Technol. A 15 (1997) 1057.
- Y.R. Park, D. Jung, Y.S. Kim, Japan J. Appl. Phys. 47 (2008) 516.
- Y.S. Cho, G.R. Yi, J.J. Hong, S.H. Jang, S.M. Yang, Thin Solid Films 515 (2006) 1864.
- Y.W. Zhu, H.Z. Zhang, X.C. Sun, S.Q. Feng, J. Xu, Q. Zhao, B. Xiang, R.M. Wang