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The Effect of the Annealing on the Properties of ZnO/Cu/ZnO Multilayer Structures

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Abstract

Study of ZnO / Cu / ZnO multilayer film stack using Cu film as a sandwich layer, in order to improve the overall performance of the transparent conductive film, structural, electrical and optical properties. ZnO / Cu / ZnO multilayer films prepared at room temperature by a DC and RF magnetron sputtering technique. The results showed that ZnO / Cu / ZnO multilayer film has good crystalline properties. With an increasing Cu layer thickness, a visible light transmittance of the multilayer film is reduced, while the electrical performance improved significantly. In the Cu layer thickness of 20 nm, ZnO / Cu / ZnO multilayer film optoelectronic integrated for optimal performance

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Keywords: RF sputtering; multilayer; Optical and Electrical properties.

1. Introduction

Ultrathin films of metals (Au, Ag, Cu) with at nanosized thickness are widely used. The transparent conductive film is a transparent and electrically conductive combination of two properties in one thin film and this kind of materials specially used in photoelectric device[1], flat panel displays, solar cell, and infrared reflective film. In Research for a long time, the transparent conductive films focused on ITO (In_2O_3:Sn) film. ITO having excellent photoelectric Performance, but also have weaknesses, such as: the price is high, toxic, and not stable enough. By Compared ITO with ZnO thin film the ZnO-based transparent conductive electric film has rich raw materials, cheap, non-toxic, etc[2]. In recent years, more possible alternatives to ITO, it's become a hot research. Currently, Al doped

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ZnO (AZO) films more widely used, but also it has a weakness because Al is a highly reactive element, thin film when the film growth Al-doped prone Al_2O_3 phase[3-5]. In order to further improve the conductivity, the ZnO-based single metal thin film is introduced with excellent conductivity. Many Conducted ZnO thin films as a multilayer structure research have been done using different metal such as Ag[6-8], Cu[9,10], Al[11,12], Fe[13,14] and so on. Metal Cu has excellent conductivity, and the price is relatively low compared with Ag and it become have more advantage.

In this study, Cu metal layer is used between two ZnO thin film layers by sputtering method at room temperature to prepare a ZnO / Cu / ZnO transparent conductive multilayer film. By design the Cu thickness to obtain high performance photovoltaic element as transparent conductive film.

2. Experimental work

Thin film growth

Figure 1 shows prepared ZnO / Cu / ZnO multilayer film structure diagram using DC and RF magnetron sputtering system at room temperature. ZnO layer and Cu layers in the order are alternately deposited on a glass substrate. On the bottom preparation ZnO film is 99.999% purity ZnO target. Sputtering power fixed at 140W. There are two layers in a multilayer film ZnO layer, each ZnO film layer sputtering time are 2min, each having a thickness of approximately 60nm.

Preparation of Cu layer with a metal purity target of 99.99% the sputtering carried out at atmosphere with the high-purity Ar gas, the gas pressure was 1.0 Pa, sputtering power is fixed at 100W. For fix Cu layer thickness we have controlled the sputtering time, which was, 5nm, 10nm, 15nm and 20nm.

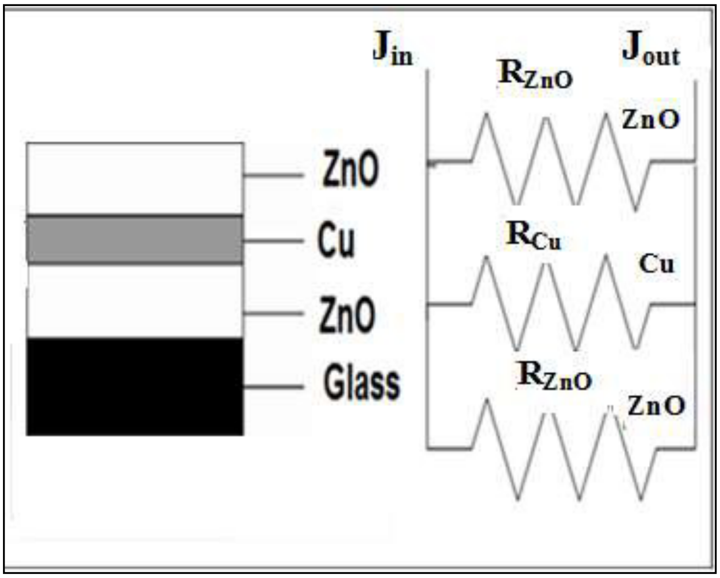

Fig.1. Schematic diagram of transparent conductive ZnO/Cu/ZnO multilayer films

3. Results and discussion

3.1. Characterization

In this study we have used XRD (PANalytical) to get the crystallization performance testing Crystal structure of the multilayer film,. Use Hall tester (Accent optical technologies Inc HL 5500pc-hall effect four-point probe method) sample Conduct resistivity, and mobility tests. Use UV- visible infrared spectrophotometer. (Agilent Technologies cary series UV-VIS-NIR sepectrophotome) for thin film sample transmission spectra test.

3.2 Structural Characteristics

Figure 2 shows a Cu layer of different thicknesses of ZnO/Cu/ZnO multilayer thin film XRD pattern of the film. It can be clearly seen from the figure to the ZnO (002) diffraction peaks, indicating that the ZnO/Cu/ZnO multilayer film, ZnO wurtzite structure, its c-axis perpendicular to the substrate surface. When the Cu layer thickness of 10nm, was observed Cu (111) peak intensity with increasing Cu layer thickness gradually enhanced.

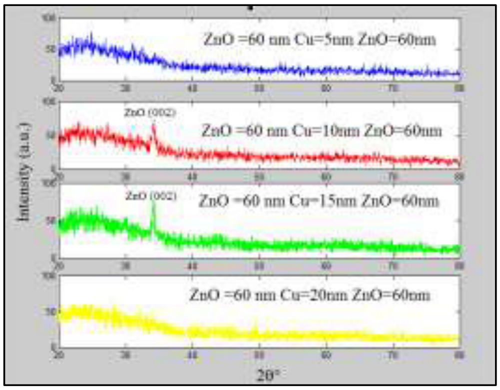

Fig. 2. XRD spectra of ZnO/Cu/ZnO multilayer films with different thicknesses of Cu layer.

Fig. 2 shows the XRD patterns of as deposited multilayer for fixed thickness of ZnO and with different thickness of Cu layer. The ZnO (002) peak intensity increasing when the Cu layer thickness increased; and with the Cu layer thickness increase, (002) peak not appeared and no peak can be observed. (002) peak intensity change by Cu/ ZnO thickness greater effect. When the Cu layer is thinner (such as 5nm) the film discontinuous structure was similar to the island, the upper layer of ZnO film growth with low crystal quality. When the Cu layer thickness to 10nm, the film becomes continuous or in semi-continuous state, sharp increase of the preferred (002) orientation of the ZnO films shows that the crystalline quality of the films is influenced by the increase of thin copper layer. But with the Cu layer thickness increases more, lead to increase in internal defects, leading to growth not quality film and (002) peak intensity decreases.

3.3. Electrical Characteristics

Figure 3 is a ZnO/Cu/ZnO multilayer film sheet resistance with different Cu layer thickness. The relationship between the Cu thickness and sheet resistance with Cu layer thickness increases from 5 nm to 10nm, the sheet resistance of ZnO/Cu/ZnO multilayer film drastically reduced from 95.1 to 89.99 Ω/sq respectively, followed by a further increase with the thickness of the Cu layer, the sheet resistance gradually decreased, when the Cu layer thickness of 20 nm ZnO/Cu/ZnO multilayer film sheet resistance was 9.877Ω /sq.

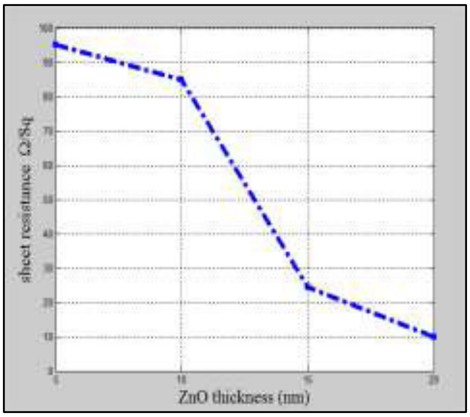

Fig.3. Variation of sheet resistance of ZnO/Cu/ZnO multilayer films with respect to the thickness of Cu layer thickness.

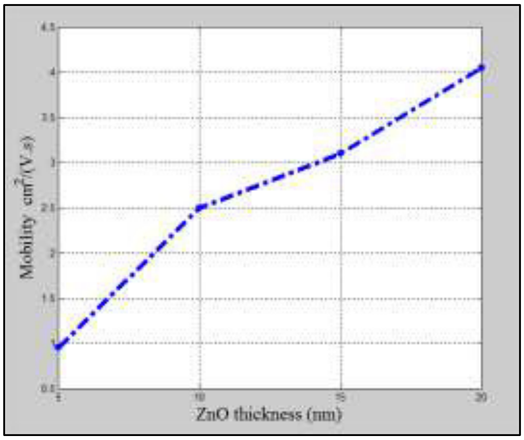

Fig.4. is a ZnO/Cu/ZnO multilayer film carrier mobility relationship with Cu thickness.

When the Cu layer thickness is thin (such as 5nm) the Cu layer is isolated like island structure[15], and the most of the current flowing from the ZnO layer, and Cu Island structure as discontinuous layer of scattering centers, so that the film mobility was very low around 0.955 cm2 / (V.s). Thereafter, with the increase in the thickness of the Cu layer, Cu layer was near like continuous state, the majority of the current starts flowing from the Cu layer, this time the mobility of the multilayer film integrated mobility on the will be rise. When the Cu layer thickness become 10nm, Cu layer films have been completed full continuous, almost all the current flows from the Cu layer and become low resistance, this mobility for multilayer film with 20nm Cu thickness was 4.05cm2 / (V · s).

The Cu metal work function is 4.5eV [16] significantly less than the work function of ZnO (Φ = 5.16eV -5.3eV) [17]. ZnO and metal band structure Cu diagram shown in fig. 5, when the Cu contact with ZnO, electrons easily injected from the Cu layer to ZnO layer. With an increasing Cu layer thickness, ZnO / Cu / ZnO a sharp increase in carrier concentration layer film. When the thickness of the Cu layer to 10nm, the multilayer film the carrier concentration increased by almost two orders of magnitude, which indicated from the value of mobility of the carriers of the multilayer structure.

Sheet resistance and resistivity was decreased but mobility was increased because the electron injection from the Cu layer to ZnO layer.

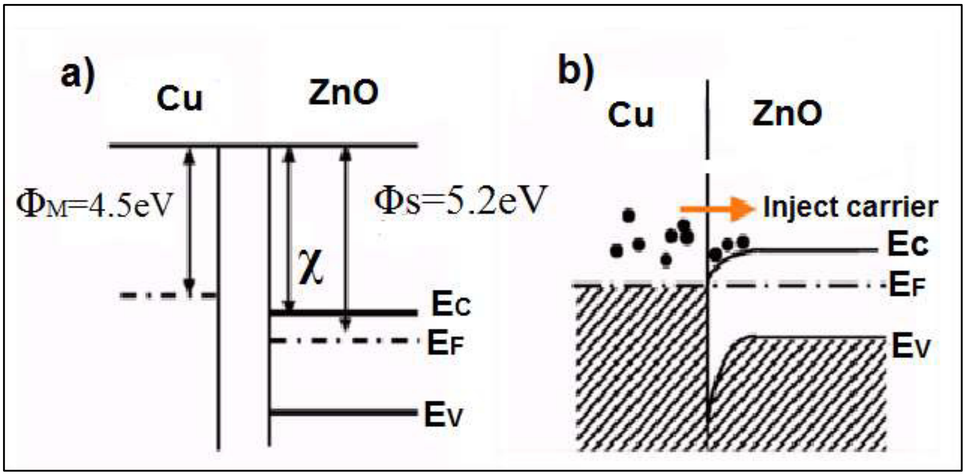

Fig.5. Schematic of band gap energy of ZnO and Cu.

3.4. Optical properties

Fig. 6 shows a ZnO with different Cu thickness multilayer thin film visible light transmission spectra. Cu as a mid-layer considers a key factor to reduce the average visible light transmittance of the multilayer film. It is seen from Fig. 6, the visible light transmittance of the multilayer film in a short wavelength and long wavelength region have a different behaviour.

However, with the Cu layer thickness increases, the transmittance decreases, because the short wavelength region is mainly affected by the transmittance of the light-absorbing effect, with the Cu Layer thickness increases more electrons to achieve band transition and absorbs more light, thereby lowering the visible light transmittance.

We can observed from Fig. 6 that the maximum transmission was happened at wavelength below 600nm for all Cu thickness [15, 18]. When the wavelengths greater than 600nm wavelength transmittance began to decrease due to long wave region, the transmittance of the film is mainly by reflectance effects and with wavelength increase reflected a significant phenomenon.

Meanwhile, when wavelengths greater than 600nm, and Cu layer thickness increased from 5nm to 10 nm, the transmittance is gradually decreased, and this is because of the larger thickness of Cu Of the multilayer film reflective become high.

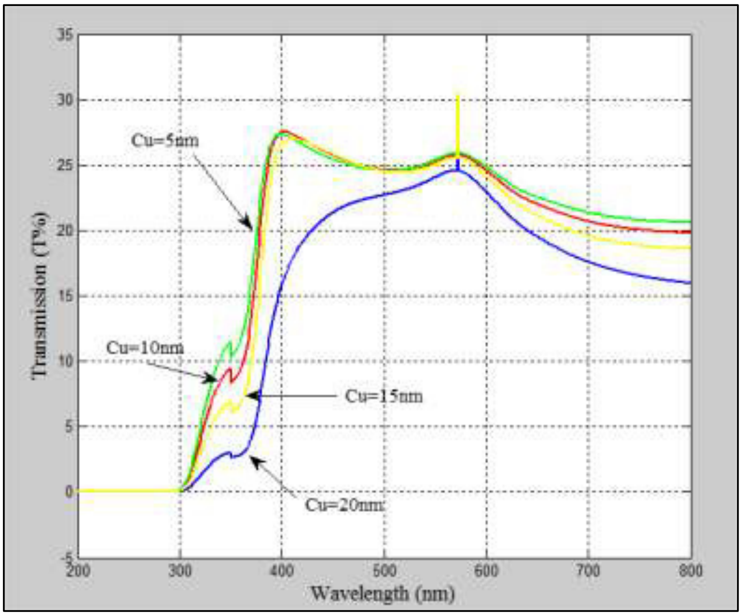

Fig.6. Transmission spectra of ZnO/Cu/ZnO multilayer films with different Cu layer thickness.

3.5. Optical properties of the multilayer structure

The Cu and ZnO thickness is a key factor of multilayer structure of a transparent conductive film. The metal layer in the stack ZnO / Cu / ZnO multilayer Conductive properties of the film increases as the thickness of the metal layer is enhanced, however, with the increase in the thickness the transparent multilayer film will be significantly reduced the transparent conductive thin film performance.

4. Conclusion

ZnO/Cu/ZnO multilayer as transparent conducting coatings have been deposited by RF and DC magnetron sputtering at room temperature.The transmittance is relatively high in the entire visible region. The results show that the presence of Cu layers decreases the visible light transmittance of the film, but at the same time enhanced the electrical properties of the film. The transmittance decreases with increasing Cu thickness in both UV and visible regions. It was found that the multilayer film can be optimized to have a low sheet resistance about 9.877 Ω/sq. and average transmittance covers 25% in the visible region.

Acknowledgements

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