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Technological Regimes Searching the Effect of Thermal Annealing on Optical and Electrical Properties of Co/ZnO Multilayer Thin Film of Different Thickness

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Abstract: The fabrication of the Co/ZnO multilayer thin film for studying the influence of thermal annealing on optical and electrical properties is presented in this paper. In this case, at first Co/ZnO multilayer films were prepared by e-beam evaporation in a vacuum at a pressure of 3.2×10^{-5} torr. In the multilayer, the thickness of Co and ZnO was kept same. Each layer thickness was varied from 5 nm to 15 nm and repeated three times. The deposition rate of the Co and ZnO thin films are about 1.33 nm/s & 1.43 nm/s respectively. The optical and electrical properties of the deposited and annealed Co/ZnO films had been studied. The average transparency of Co/ZnO multilayer thin film is roughly about 55% and decreased with increasing film thickness and increased when annealed. The T. C. R. of deposited and annealed Co/ZnO multilayer thin films in all cases is negative which indicates that the thin films are semiconducting in nature.

Keywords: Co, ZnO, E-Beam, Multilayer, Optical Properties, Electrical Properties, Thickness

1. Introduction

Dilute magnetic semiconductors (DMSs)—semiconductors doped with a few percent of magnetic atoms—have been intensely researched in the past few years due to their promising application in spintronics devices [1]. Based on the theoretical works of Dietl et al. [2], ZnO-based DMS has attracted a considerable amount of interest due to their proposed applications of room-temperature ferromagnetism. In recent years, with the evolution of spintronics, transitional metal doped ZnO thin film have found potential applications in spintronics and photonics devices [3, 4]. A powerful review of DMS state of the art by Pearton et. al. [19] summarized that ZnO: Co system has particularly promising applications in spintronics that requires ferromagnetism near room temperature. Risbud et. al. [20] also suggested that the wide bandgap wurtzite phase semiconductor ZnO is very suitable to be the matrix in DMS as the zinc ions can be substituted by magnetic transition metal ions to yield a metastable solid solution. Nevertheless, zinc oxide is also optically transparent and an n-type semiconductor. Chambers et. al. [21] stated that n-type is attractive for spintronics because electrons exhibit longer spin relaxation time than holes. Zinc oxide is also a direct band gap semiconductor with Eg = 3.35 eV at room temperature [22]. ZnO is also a material where the doping system is always having a Tc well above room temperature. Therefore, it is interesting to study the influence of thermal annealing on optical and electrical properties of Co/ZnO multilayer thin film. Many studies described the fabrication techniques of ZnO thin film [4-11]. However, Co doped ZnO thin film has been deposited by Pulsed Laser Deposition (PLD), Molecular Beam Epitaxy and Sputtering methods [12-18]. Considering the film purity and quality electron beam evaporation has been adopted as a noble and widely used film fabrication technique.

2. Experimental Setup

The Co/ZnO multilayer thin films were prepared by e-beam evaporation method (Edward-306) in a vacuum at a pressure of 3.2×10^{-5} torr on ultrasonically cleaned glass substrates. The thickness of Co and ZnO was kept same. Optical interference method is used to measure the thickness of Co and ZnO films. Each layer thickness was varied from 5 nm to 15 nm and repeated three times. The sample size was 10 mm × 10 mm. There were three types of films. S1: [Co(5 nm)/ZnO(5 nm)]×3, S2: [Co(10 nm)/ZnO(10 nm)]×3, and S3: [Co(15 nm)/ZnO(15 nm)]×3. A two-stage process has been used to prepare Co/ ZnO multilayer thin films. Firstly Co thin film has been deposited onto glass substrate at deposition rate 1.33 nm/s and then ZnO thin film has been deposited on the resulting Co layer at deposition rate 1.43 nm/s and repeated three times to produce Co/ZnO multilayer thin film and the upper layer was ZnO. The films were annealed in an oven in open air for an hour in the temperature 773K. Optical study has been carried out by SHIMADZU UV-visible spectrophotometer (UV-1650PC). Electrical measurements were carried out using Vander-pauw technique [23].

3. Results and Discussion

3.1. Optical Properties

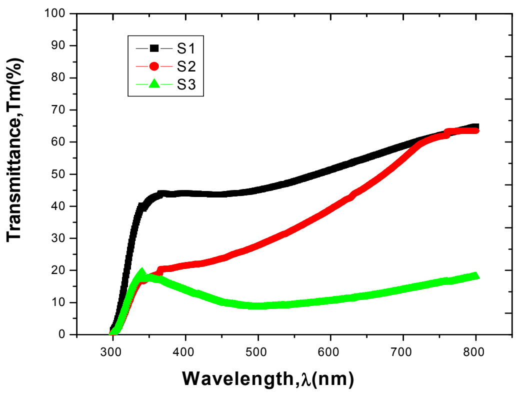

Figure 1. *Variation of transmittance with wavelength for the as-deposited Co/ZnO multilayer thin films of different thicknesses.*

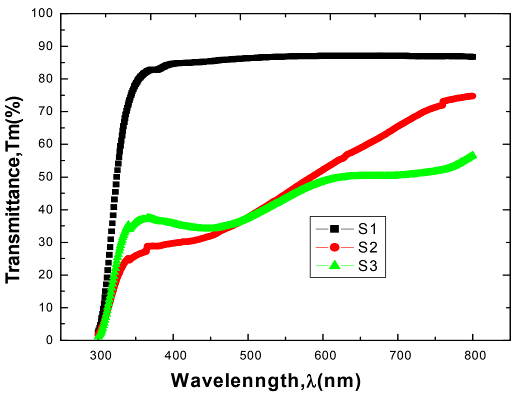

Figure 2. *Variation of transmittance with wavelength for the Co/ZnO multilayer thin films of different thicknesses annealed at temperature 773K.*

The optical transmittance has been measured for the as-deposited and annealed Co/ZnO film by the spectrophotometer in the wavelength range of 300-800 nm. Figure 1 shows the transmittance spectra of as deposited and Figure 2 shows the transmittance spectra of Co/ZnO multilayer films annealed at 773K for an hour. It is observed that the transmittance of all these films is nearly zero in 300 nm and the lower thickness film has higher transmittance than the higher thickness film. Interestingly, it is observed that the transmittance is increased at the same thickness when the film is annealed. This increase in transmittance might be due to the interdiffusion and oxygen adsorption in the films. In the case of S3 film annealed at 773 K temperature it shows the wavelike nature. This nature may occur due to two reasons, one is roughness of the thin film and another is multiple reflections [24-26] from different interfaces. As the annealing temperature increases roughness of the surfaces also increases which increases the wavelike nature. Because the grain size increases with the increase in the annealing temperatures, due to reduction of grain boundaries in ZnO thin film [27]. Multiple reflections may occur due to the creation of the diffused layer of Co and ZnO in each interface.

3.2. Electrical Properties

The temperature dependent transport plays an important role in thin film characterization. To measure TCR at first the voltage and current of the sample have been measured for different temperatures. After collected these data the resistivity of the films for different temperatures have been calculated using Van-der- Pauw's method.

According to the law of Ghosh and Deb [28] the relation between resistivity and temperature is given by

$$\rho_T=\rho_0(1+\alpha T+\beta T^2+\dots\dots\dots) \quad (1)$$

where, ρ_0 is a constant ρ_T is the resistivity at T K. α is the temperature coefficient of resistance (TCR) and β is a constant. Generally $\alpha>>\beta$ at low temperature, and we can write from first approximation

$$\rho_T=\rho_0(1+\alpha T) \quad (2)$$

At temperature T_2 anT_1 equation (2) can be written as

$\rho_{T2} = \rho_0\ (1+\alpha T_2)$ and

$\rho_{T1}= \rho_0\ (1+\alpha T_1)$

Using these two equations we can write

$$\alpha=\frac{(\rho_{T2}-\rho_{T1})}{\rho_{T1}(T_2-T_1)} \quad (3)$$

Generally ρ_{T1} and ρ_{T2} are the resistivity at temperatures T_1 and T_2.

Then the temperature coefficient of resistance (TCR) is estimated from the measured data according to the equation (3). Figure 3 and Figure 4 depict the TCR of as-deposited and annealed Co/ZnO thin films of various thicknesses annealed at 773K for an hour. It is observed that TCR is negative in all cases. This indicates that the films are semiconducting in nature. However, TCR does not change systematically which is difficult to explain. The interdiffusion of the metal Co particles in insulating ZnO layers can make complex hopping transport mechanism. Ionized impurities are the important source of scattering in doped semiconductor and if this dominates the TCR can become negative.

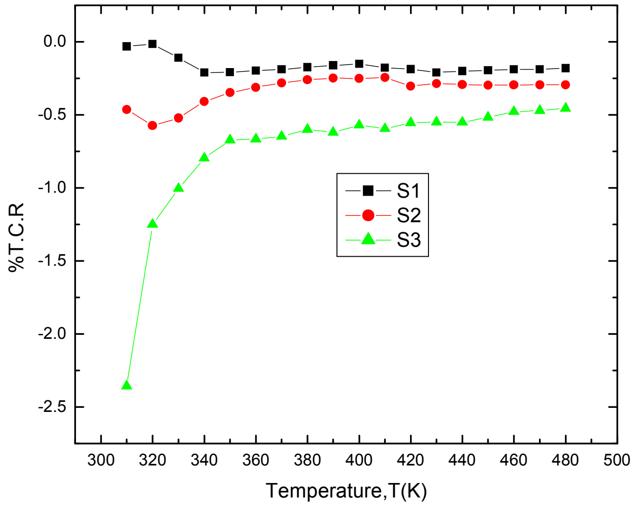

Figure 3. *Variation of T. C. R. with temperature for the as-deposited Co/ZnO multilayer thin film of different thicknesses.*

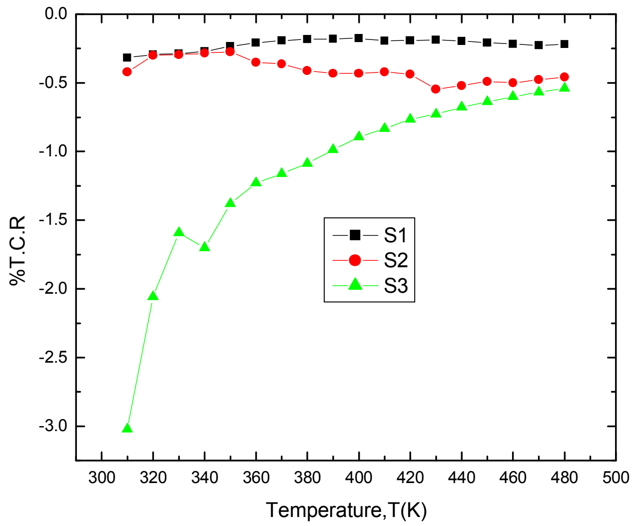

Figure 4. *Variation of T. C. R. with temperature for the Co/ZnO multilayer thin film of different thicknesses annealed at temperature 773K.*

4. Conclusions

The temperature dependent optical and electrical properties of Co/ZnO multilayer thin films have been studied. It is concluded from the experimental result that the effect of annealing plays an important role in the optical and electrical properties of the films. The as-deposited Co/ZnO multilayer thin films show low transmittance in the visible range. The post annealing at moderate temperature (773K) can give higher transparency in the visible range. The average transmittance is obtained for thinner sample (S1) is nearly 70%. The annealing at higher temperature increases the transmittance. As-deposited and annealed sample of Co/ZnO shows the negative T. C. R. which indicates that the films are semiconducting in nature. The key property for obtaining transparent magnetic oxides material for future multifunctional material is that it should exhibit magnetic, semiconducting property and at the same time it should be highly transparent. The role of annealing in Co/ZnO multilayer thin films gives the path to control the electrical and optical properties.

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