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The Effect of Inter diffusion on The Electrical And Optical Properties of Co/TiO₂ Multilayer Thin Films

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Abstract: The Co/TiO2 multilayer thin films were fabricated by e-beam evaporation technique on glass substrate. The electrical and optical properties of as-deposited and annealed Co/TiO₂ multilayer thin films were studied. The interdiffusion of Co and TiO₂ plays dramatic change in the electrical conductivity and optical transmittance of the film. The Co/TiO₂ multilayer thin films annealed at 473K exhibit semiconducting behavior and at the same time it possess about 70% of transmittance value which is much larger compare to the as-deposited one.

Keywords: Transparent magnetic oxides, Diluted magnetic semiconductor, Interdiffusion, Multiple reflection etc.

1 Introduction

Transparent magnetic oxides have a high potentiality for fabricating future multifunctional spintronic devices according to the various technological reports and roadmaps [1-5]. One of the approaches in achieving this goal is the doping of transition magnetic (TM) material into transparent oxide [4-6]. In several cases, TM doped oxide such as Co doped ZnO, TiO2, SnO2 etc. shows ferromagnetism at room temperature [5-12]. The mechanism and origin of ferromagnetism and its reproducibility is still studied. Among the various oxides, Co doped TiO2 is very attractive and one of the most prominent material [3,7]. This material shows ferromagnetism at room temperature, and it has a higher Curie temperature [3]. The requisite property of transparent magnetic oxides to use it as multifunctional is to obtain larger magnetization, higher optical transmittance as well as semiconducting behavior. In this paper, we would like to investigate how Co diffusion can change the electrical and optical properties. After the preparation of the Co/TiO2 multilayer, annealing at high temperature can cause interdiffusion of Co into TiO2 or vice versa. The interdiffusion can be controlled by the annealing temperature and annealing time. The conduction mechanism is quite complex in the metal doped oxide materials [13]. There are few reports on the effect of interdiffusion on the electrical and optical properties of Co/TiO2 multilayer thin films. In

this paper, the electrical and optical properties of ${\rm Co}/{\rm TiO_2}$ multilayer thin films of as-deposited as well as annealed films have discussed.

2 Experimental

The Co/TiO2 multilayer thin films were prepared by e-beam evaporation method (Edward-306) in a vacuum better than 10-5 Torr on glass slide substrate. The thickness of Co and TiO2 was kept same. Each layer thickness was varied from 5nm to 15 nm and repeated three times. The sample size was 5 mm \times 5 mm. There were three types of films. S1: Co(5nm)/TiO2(5nm)×3, S2: Co(10 nm)/TiO2(10 nm)×3, and S3: Co(15 nm)/TiO2(15 nm)×3. The deposition rate of the Co and TiO2 thin films were about 1.33 nm/sec &.1.25 nm/sec respectively. The films were annealed in an oven in the temperature range of 373K-773K. Atomic force microscopy (AFM) has been used to characterize the surface morphology (XE-70, Park Systems). The optical transmittance, T have been measured in the wavelength range from 200 to 800nm using a UV-Visible (SHIMADZU UV1650PC) double beam spectrophotometer.

3 Results And Discussion

3.1 Electrical Properties

Electrical measurements were carried out using Van der Pauw technique [14]. The conductivity was calculated from the measured data. Fig. 1 shows the conductivity of asdeposited and annealed Co/TiO₂ (S1) thin films. It is in the order of $10^4(\Omega-m)^{-1}$ which is in the semiconductor range. The conductivity is found to increase with temperature because the number of carriers available for electrical conduction is increased with temperature [13]. It is noteworthy to mention that Co/TiO₂ multilayer thin films were post annealed at different temperatures in open air for one hour. It shows that the post annealing decreases the electrical conductivity of the films. Annealing might cause atomic interdiffusion or nanoparticle formation (the island-like structures on the AFM image of "473K" annealed sample (Fig. 5)) or both of Co and TiO₂. The atomic interdiffusion or nanoparticle formation or both might reduce the effective conductive channel width. This results the decrement of conductivity.

The temperature dependent transport plays an important role in thin film characterization. The temperature coefficient of resistance (TCR) is estimated from the measured data according to the formula reported elsewhere [13]. Fig. 2 depicts the TCR of Co/TiO₂ thin films of various thicknesses annealed at 673K. It is observed that TCR is negative in all cases. This indicates that the film are semiconducting in nature. However, TCR does not change systematically which is difficult to explain. The interdiffusion of the metal Co particles in insulating TiO₂ 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

3.2 Optical Properties

The optical transmittance have been measured for the asdeposited TiO₂ (30nm), Co (30nm) and the as-deposited and annealed Co/TiO_2 (S1) film by the spectrophotometer in the wavelength range of 200-800 nm. Fig. 3 shows the transmittance spectra of those films. It is observed that the transmittance of all these films is nearly zero in the UV range (below 300 nm). The TiO₂ thin film is obtained highly transparent in the wavelength range of 350 to 800 nm and the transmittance value is nearly constant in the entire range. The average transmittance in this range is about 87% which is comparable to the other reports [7]. On the other hand, the Co film shows low transmittance in same wavelength range (350-800 nm). The average transmittance in this range is obtained about 35%. However, a slight increase of transmittance at longer wavelength is noticed in this case. The transmittance spectrum of as-deposited Co/TiO₂ films is nearly the same as the Co film. The presence of Co reduce the transmittance of the Co/TiO₂ thin film. Interestingly, it is observed that the transmittance is increased of the annealed films compare to the as-deposited one. This increase in transmittance might be due to the interdiffusion and oxygen adsorption in the films. This is the same reason as in the case of the reduction in conductivity. The average transmittance of S1 film annealed at 473K is about 70%. The transmittance is found to increase with the wavelength. However, annealed with higher temperature 673K, the transmittance (as shown in fig. 3), the transmittance is found to reduce slightly and a small oscillation is observed in the transmittance spectra. This oscillation might be due to the interference which causes multiple reflections in the films. Fig. 4 shows the transmittance spectra of Co/TiO₂ thin film of different thickness annealed at 473K. It is observed that thinner sample show higher transmittance value which is simply the effect of thickness.

3.3 Surface Morphology

The surface morphology of the as-deposited and annealed Co/TiO2 thin films was studied using Atomic Force Microscopy. Fig. 5 depicts the AFM images of as-deposited, annealed (at 473K and 673K) Co/TiO2 thin films. It is obtained from the measurement that the sample roughness in all cases is less than 1 nm. However, the sample roughness is little higher in annealed case compare to the as-deposited one.

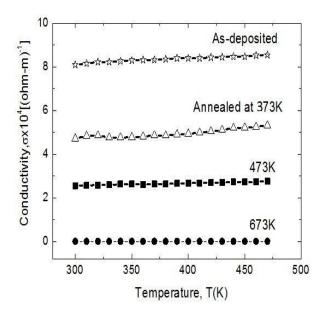


Fig.1 The electrical conductivity as a function of temperature of the as-deposited and annealed Co/TiO_2 (S1) multilayer thin film grown on glass substrate.



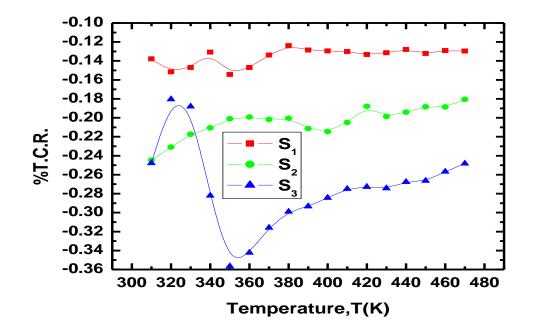


Fig.2 Variation of T.C.R. with temperature for the Co/TiO_2 multilayer thin film of different thicknesses annealed at 673K.

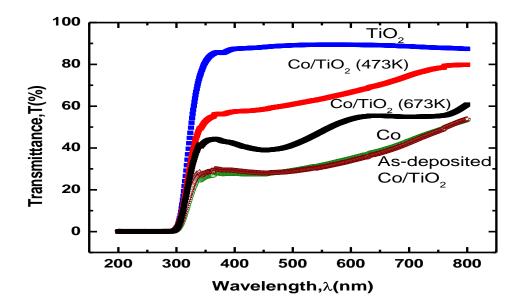


Fig.3 The transmittance spectra for the as-deposited Co, TiO_2 , Co/TiO_2 and annealed Co/TiO_2 multilayer thin films (S1) grown on glass substrate.



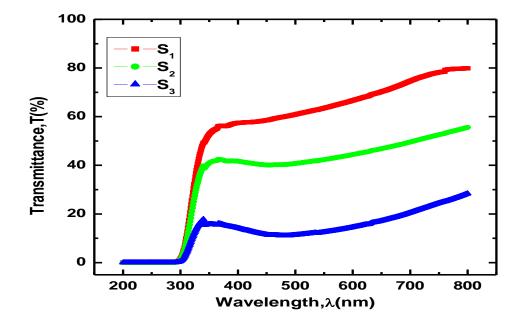


Fig.4 Variation of transmittance with wavelength for the Co/TiO_2 multilayer thin films of different thicknesses annealed at temperature 473K.

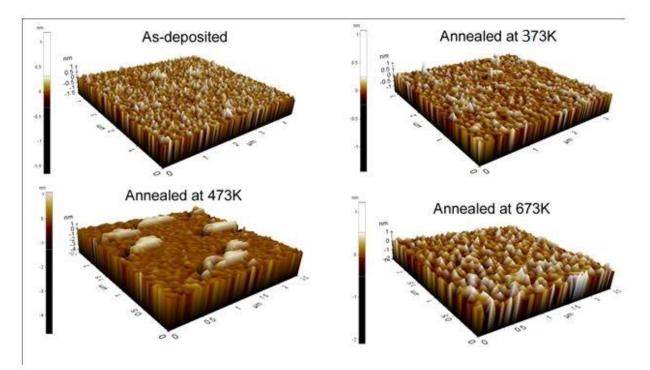


Fig.5 The AFM images of as-deposited and annealed Co/TiO2 multilayer thin film (S1) annealed at different temperatures.

4 Conclusions

The temperature dependent electrical and optical properties of Co/TiO₂ multilayer thin films have been studied. It is concluded from the experimental result that the effect of interdiffusion plays an important role in the electrical and optical properties of the films. The as-deposited Co/TiO₂ thin film show higher conductivity and low transmittance in the visible range. The post annealing at moderate temperature (473K) can give semiconducting behavior as well higher transparency in the visible range. The room temperature conductivity of Co/TiO2 films annealed at 473K grown on glass substrate is obtained in the order of $10^4(\Omega$ m)⁻¹ and the average transmittance is obtained for thinner sample (S1) is nearly 70%. The annealing at higher temperature reduces the conductivity as well as transmittance. 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 interdiffusion in Co/TiO2 multilayer thin films in the electrical and optical properties gives not only the path for obtaining the requisite properties in the application point of view but also gives interesting underlying physics for fundamental research.

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