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Study Electrical and Optical Properties of Co/TiO_2 Multilayer Thin Films

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Abstract— The Co/TiO_2 multilayer thin films were prepared by e-beam evaporation on glass substrate. Each film were consist of three alternative bi-layer of Co and TiO_2 and the top layer of the films were TiO_2. Films were annealed in open air at different temperatures. Temperature dependence of conductivity was measured for all the samples. It was observed that conductivity was found to increase with temperature. The conductivity of as-deposited film was higher than the conductivity of annealed film. Thickness dependence of conductivity was also studied. It was also found that, as the film thickness increased, conductivity decreased. The conductivity of the annealed film was of the order of $10^2(\Omega m)^{-1}$. The T.C.R of Co/TiO_2 multilayer thin films was negative for both as deposited and annealed films. All those results suggest that Co/ TiO_2 multilayer annealed thin films are semiconducting in nature.

The optical transmittance was measured for as-deposited and annealed samples. It was obtained that the annealing samples exhibit higher transmittance compare to the as-deposited one. At same film thickness, transmittance increased with annealed temperature increased. Thus annealing plays an important role for the change in electrical and optical properties. In summery we have seen that Co/TiO_2 multilayer thin films are semiconducting in nature and optically transparent.

Keyword's—**Transparent magnetic oxides, Diluted magnetic semiconductor, Interdiffusion, Multiple reflection etc.**

I. 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, TiO_2 and SnO_2 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 TiO_2 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. Although, there are some works in Co doped TiO_2, there are few works on the electrical and properties of Co/TiO_2 multilayer thin films. The conduction mechanism is quite complex in the metal doped oxide materials [13]. In this paper, we would like to investigate how annealing plays a role in the electrical and optical properties of Co/TiO_2 multilayer thin films. After the preparation of the Co/TiO_2 multilayer, the samples were annealed at high temperature and the electrical and optical properties were compared of Co/TiO_2 multilayer thin films of as-deposited with the annealed films.

II. Experimental

The Co/TiO_2 multilayer thin films were prepared by e-beam evaporation method (Edward-306) in a vacuum better than 10^{-5} Torr on glass substrate. The thickness of Co and TiO_2 was kept same. Optical interference method is used to measure the thickness of Co and TiO_2 films. Each layer thickness was varied from 2 nm to 4 nm and repeated three times. The sample size was 5 mm × 5 mm. There were three types of films. S1: $[Co(2nm)/TiO_2(2nm)]_{\times 3}$, S2: $[Co(3\ nm)/TiO_2(3\ nm)]_{\times 3}$, and S3: $[Co(4\ nm)/TiO_2(4\ nm)]_{\times 3}$. The deposition rate of the Co and TiO_2 thin films were about 1.33 nm/sec &.1.25 nm/sec respectively. The films were annealed in an oven in

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open air in the temperature range of 473K-673K. The optical transmittance, ***T*** have been measured in the wavelength range from 200 to 800nm using a UV-Visible (SHIMADZU UV1650PC) double beam spectrophotometer.

III. Result and Discussion

A. Electrical Properties

Electrical measurements were carried out using Vander-pauw technique [14]. The conductivity was calculated from the measured data. Figure 1 shows the conductivity of as-deposited and annealed Co/TiO_2 (S2) thin films and Figure 2 shows the conductivity of as-deposited and annealed Co/TiO_2 (S3) thin films. For both case, the conductivity is in the order of $10^2(\Omega\text{-m})^{-1}$ 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/TiO_2 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. Although there is no direct evidence, however, there is a possibility that annealing cause interdiffusion of Co and TiO_2. The interdiffusion might reduce the effective conductive channel width.

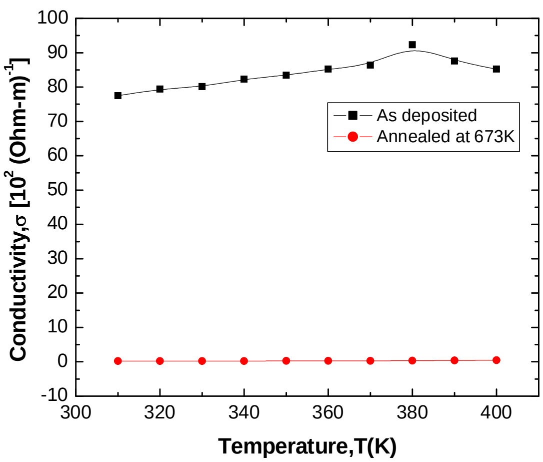

Fig. 1: Variation of conductivity with temperature of S2 film.

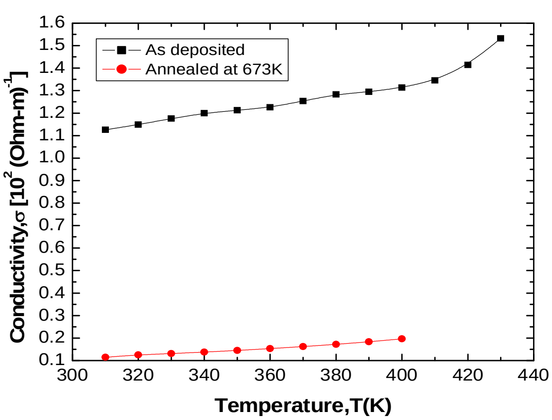

Fig. 2: Variation of conductivity with temperature of S3 film

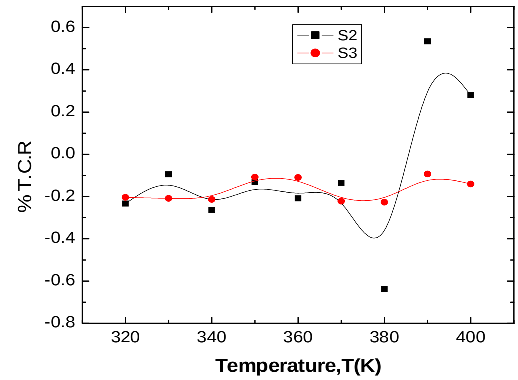

Fig. 3: Variation of T.C.R with temperature of as deposited films of different thickness.

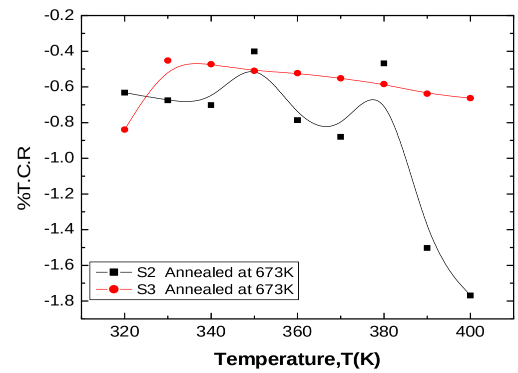

Fig. 4: Variation of T.C.R with temperature of annealed [673K] films of different thicknes.

This results the decrement of conductivity. It is also observed that higher thickness Co/TiO_2 thin films show lower conductivity compare to the lower thickness one for both as-deposited and annealed.

The room temperature conductivity of as-deposited S2 film was $77.5\times10^2(\Omega m)^{-1}$ while the room temperature conductivity of as-deposited S3 film was $1.125\times10^2(\Omega m)^{-1}$. For annealed film, the conductivity of S2 film was $0.1915\times10^2(\Omega m)^{-1}$ while the conductivity of S3 film was $0.119\times10^2(\Omega m)^{-1}$.

In Figure 1, for annealed film, the increase of conductivity with temperature is very small and because of this the curve looks like a straight line.

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]. Figure 3 and 4 depict the TCR of as-deposited and annealed Co/TiO_2 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 TiO_2 layers can make complex hopping transport mechanism.

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Ionized impurities are the important source of scattering in doped semiconductor and if this dominates the TCR can become negative.

It is noted that the S1: $[Co(2nm)/TiO_2(2nm)]_{\times 3}$ film was very thin and oxidation may result discontinuity of the film. The result of S1 film is not included as we were not able to measure correctly the electrical conductivity of the films using Van-der-Pauw technique.

B. Optical Properties

The optical transmittance has been measured for the as-deposited and annealed Co/TiO_2 film by the spectrophotometer in the wavelength range of 200-800 nm. Figure 5 shows the transmittance spectra of Co/TiO_2 films annealed at 473K.

It is observed that the transmittance of all these films is nearly zero in the UV range (below 300 nm). It is also observed that the lower thickness film has higher transmittance than the higher thickness film. Figure 6 shows the variation of transmittance with wavelength of Co/TiO_2 film annealed at different temperature. Interestingly, it is observed that the transmittance is increased of the higher annealed film compare to the lower annealed one at the same thickness.

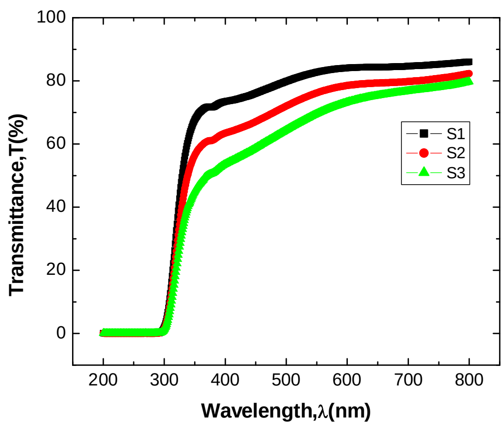

Fig. 5: Variation of transmittance with wavelength for the Co/TiO2 multilayer thin films of different thickness annealed at 473K .

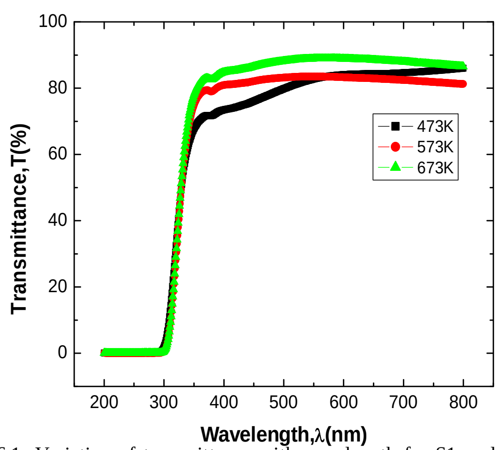

Fig. 6.1: Variation of transmittance with wavelength for S1 multilayer thin film annealed at different temperature.

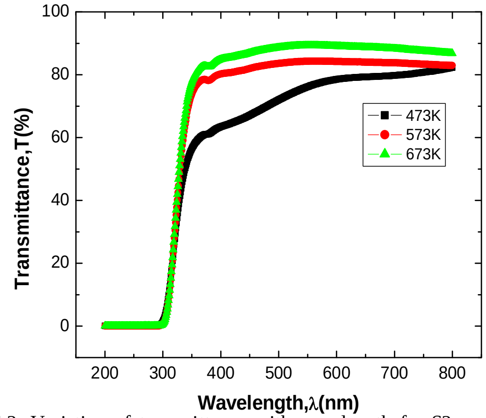

Fig. 6.2: Variation of transmittance with wavelength for S2 multilayer thin film annealed at different temperature.

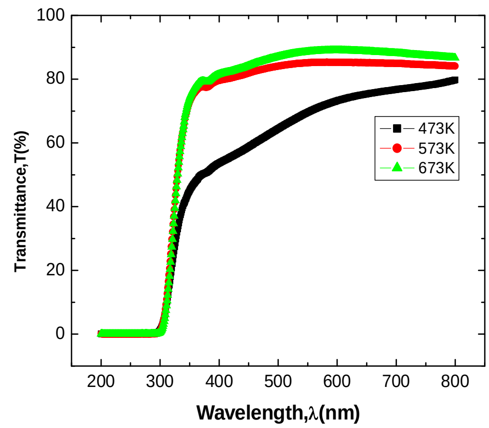

Fig. 6.3: Variation of transmittance with wavelength for S3 multilayer thin film annealed at different temperature.

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%, annealed at 573k is about 80% and annealed at 673K is about 85%.The average transmittance of S2 film annealed at 473K is about 60%, annealed at 573K is about 78% and annealed at 673K is about 83%.The average transmittance of S3 film annealed at 473K is about 50%, annealed at 573K is about 77% and annealed at 673K is about 80%.The transmittance is found to increase with the wavelength. However a small oscillation is observed in the transmittance spectra. This oscillation might be due to the interference which causes multiple reflections in the films.

IV. Conclusion

The temperature dependent electrical and optical properties of Co/TiO_2 thin films have been studied. It is concluded from the experimental result that the effect of annealing plays an important role in the electrical and optical properties of the films. The as-deposited Co/TiO_2 thin films show higher conductivity and low transmittance in the visible range. The

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post annealing at moderate temperature (473K) can give semiconducting behavior as well higher transparency in the visible range. The room temperature conductivity of Co/TiO_2 films annealed at 473K grown on glass substrate is obtained in the order of $10^2(\Omega\text{-m})^{-1}$ and the average transmittance is obtained for thinner sample (S1) is nearly 70%. The annealing at higher temperature reduces the conductivity but increase the 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 annealing in Co/TiO_2 multilayer thin films gives the path to control the electrical and optical properties.

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