Opto-Electronic Characterization of TiO_2/Metal/TiO_2 Multilayers

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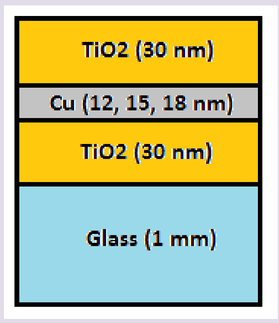
Introduction

Transparent conductive oxides (TCOs) play an important role in many optoelectronic devices such as solar cells, organic light emitting diodes, touch panel and efficiency energy applications, heat mirrors and thermoelectric materials [1]. In recent years, many researchers proposed a TCO/metal/TCO multilayer structure with advanced electrical properties, mechanical flexibility, chemical stability and high optical transparency than a TCO single layer [2]. Multilayer films of *TiO_2/Cu/TiO_2* and *TiO_2/Co/TiO_2* were grown on glass substrate by DC magnetron sputtering technique at low temperature deposition for low cost applications. Copper interlayer was chosen as a possible replacement of Ag due to their comparable electrical conductivity and relativity low cost. Cobalt interlayer is studied for ferromagnetic properties at room temperature because of its potential applications in spintronics. The deposition time was chosen to obtain estimated thickness for layers of 30 nm while metal interlayers of 12, 15 and 18 nm thick were grown. The optical, electrical, structural and morphological properties of these films were characterized by UV-visible spectroscopy, four probe technique, X-rays diffraction patterns and atomic force microscopy (AFM) respectively. Figures of merit (FOM) are determinate for transparent solar cells electrode and others applications.

Method and Materials

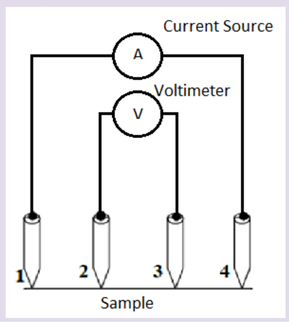
Multilayer films *TiO_2/Metal/TiO_2* of different metal (Co, Cu) thicknesses were deposited on commercial glass substrate by DC magnetron co-sputtering deposition system (ATC ORION 8HV AJA International Corporation) using metallic Ti, Cu and Co targets (99.99 % purity, 2 inch diameter, 5 mm thick, ACI alloys Inc).



Optical transmission characterization was also performed at room temperature with a Hamamatsu L2175 UV–VIS spectrophotometer in the 300 to 850 nm wavelength range (lamp 150 W).

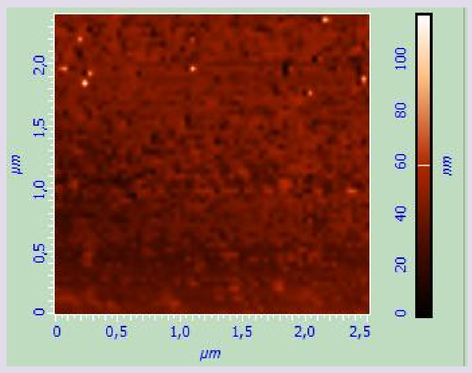
Surface voltage measurements were done using a NT-MDT atomic force microscope (AFM) in atmospheric conditions. A Si (n-type) cantilever coated with (APP NANO) is used. The probe operates at a resonant frequency of 300 KHz, Q factor of 280 and a spring constant k of 40N/m.

The *electrical resistivity* was measured using the four probe method. Electrical current was provided by a Keithley 2400 Source Meter, and the voltage was measured using a Keithley 2001 Multimeter.



Structural and Morphological Characterization

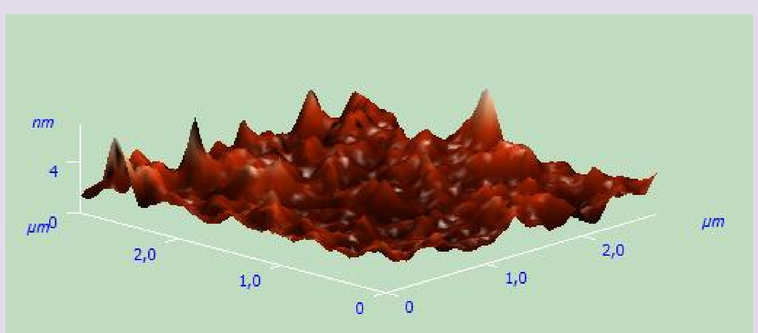
2D-3D AFM surface topography 10 x 10 for Cu and Co multilayers with different metal thickness were obtained. Average roughness analyses were carried out by NOVA software. Henceforth, as the Cu interlayer thickness increases, average roughness decreases while for Co a clear tendency is not observed. X-ray diffraction patterns of all multilayers are amorphous.



Thickness (nm)	Cu	Co
12	6,15	2,44
15	5,83	1,08
18	1,67	6,30

2D topography TiO_2/Co (15 nm)/TiO_2

AFM average roughness

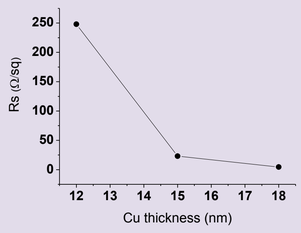


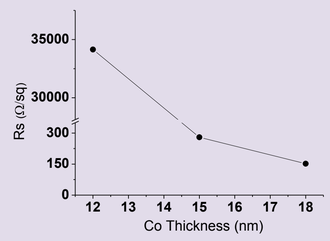
3D topography TiO_2/Cu(18 nm)/TiO_2

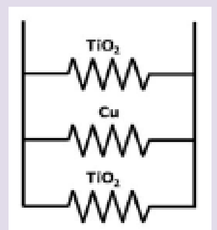
Electrical Characterization

By using the four-point probe (4P) method, the sheet resistance (Rs) multilayers can be calculated. Effects of metal interlayer thickness on Rs multilayer without annealing are shown below:

$$R_S = \frac{\pi}{ln2}\frac{V}{I}$$



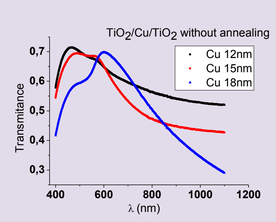


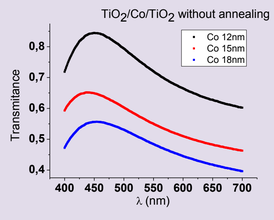


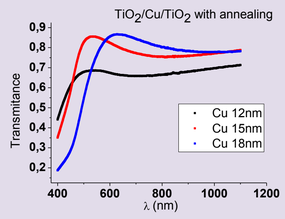
• All annealed multilayers show R sheet more than 50 MΩ.

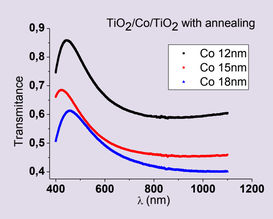
Optical Characterization

The obtained spectra display a typical behavior with a well defined absorption band edge. As metal interlayer thickness increases, a decrease in transmittance at long wavelength region is observed due to the availability of more bound electrons (Drude model).







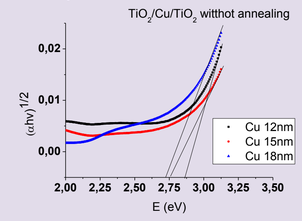


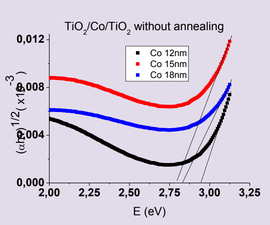
Thickness (nm)	Cu	Cu(a)	Co	Co(a)
12	65	65	69	70
15	63	75	53	54
18	61	69	46	50

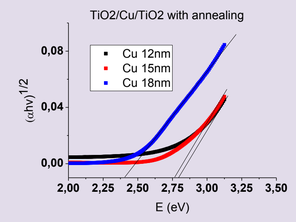
Table I: Av. T (400-800 nm)

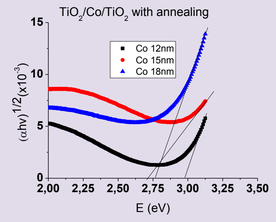
In order to examine the optical and electrical properties of multilayers films annealings at 150 C in air for 15 min were done. For Cu interlayer a clear increase in the transmittance is observed after the annealing process. Instead no major effect results for Co.

Tauc plots for indirect allow transition are shown below for all multilayers.









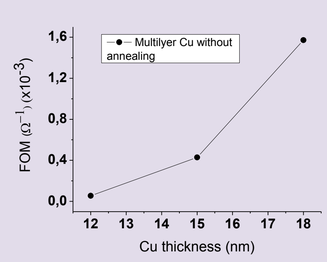
Thickness (nm)	Cu	Cu(a)	Co	Co(a)
12	2,72	2,4	2,8	3,0
15	2,75	2,7	2,83	2,7
18	2,86	2,8	2,95	2,73

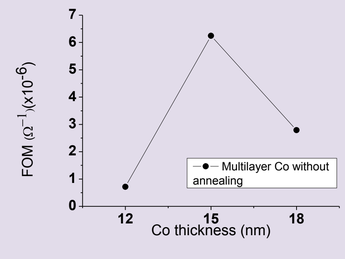
Table-II: Optical band gap (eV)

Figure of merit of TCO multilayer

The figure of merit (Haacke) of all multilayer (without annealing) are:

$$F_{TCO} = \frac{T^{10}}{R_S}$$





Conclusion

• *TiO_2/Metal/TiO_2* multilayers with different interlayer thickness (metal=Cu, Co) were fabricated by DC reactive magnetron sputtering for opto-electronic application.

• The resistivity shows a similar behavior for both metal although Co layers display higher resistivity values.

• The optical properties were analyzed as a function of metal thickness and heat treatments being their behavior different for each metal.

• For all annealed samples a slight diminution on reflectivity (high T) at NIR region is obtained while a high electrical resistivity is observed.

• The best figure of merit (*FOM,* defined by Haacke) of multilayers films without annealing were obtained for Cu 18 nm thickness and for Co 15 nm.

References

[1] Stadler, A. "Transparent conducting oxides. An up to date overview". *Materials* 5, 661-683 (2012).
[2] Dhar and T. L. Alford, "High quality transparent TiO_2/Ag/TiO_2 composite electrode films deposited on flexible substrate at room temperature by sputtering" *Appl Materials* 1, 012102 (2013).