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A study on the dependence of DC electrical properties and nanostructure of Cu thin films on film thickness

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ABSTRACT

This paper reports the correlation between film thickness, nanostructure and DC electrical properties of copper thin films deposited by PVD method on glass substrate. X-ray diffraction (XRD) and atomic force microscopy (AFM) were used for crystallography and morphology investigation, respectively. Resistivity was measured by four point probe instrument, while a Hall effects measurement system was employed for Hall Effect analysis. The grain size calculated from XRD and AFM, roughness, resistivity, hall coefficient, carrier concentration and mobility were plotted as a function of thickness. The result showed amorphous structure for 20 nm thickness, but with increasing the film thickness, Cu(111) preferred orientation was observed. The grain size, roughness and concentration of carriers increased and resistivity, hall coefficient and mobility decreased with increasing the film thickness. The result of copper thin films electrical investigation showed the value of resistivity and concentrations of carriers come to bulk state value at approximately 160 nm thickness.

Keywords: *Thin film; Copper; Thickness; Nanostructure; Resistivity; Concentration of carriers.*

INTRODUCTION

Growth of thin metal films and the relation of their different properties with deposition parameters has become a subject of great interest to those using physical vapor-deposition processes. The reason is that their structure and related properties can be modified and controlled as a function of deposition parameters and preparation method. Copper films are promising and used in many electronic devices and technology for high electrical and thermal conductivity and their relatively high melting temperature [1–4]. Furthermore, Noble metal, such as Cu thin film on transparent insulating substrate (i.e., SiO) is technologically important in the power-supply electrodes for display included integrated electron emitter [5].

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Cu has become the choice of the next generation interconnection material replacing Al, too [6]. Copper has strong advantages such as lower resistivity, superior resistance to electromigration, and stress voiding phenomena compared to the aluminum alloys. The room temperature resistivity of Cu (1.67 μΩ.cm) is approximately 40% lower than that of Al (2.7 μΩ.cm) and 50% lower than Al alloys such as Al–Cu (0.5 at.%) [6].

Many researchers have reported on the production of copper thin films, using different methods, while they also modified these films properties (such as electrical, optical and mechanical properties) as function of deposition parameters [6-14]. In this work, among the modern methods of thin film preparation, we have used physical vapor deposition (PVD) to deposit copper thin films of different thickness, and have investigated the dependence of film resistivity and nanostructure on film thickness. Furthermore, Hall effect of samples was studied as a function of film thickness. The Hall's effect is important because it enables us to make measures of mobility and concentration, and gives insight into the mechanism of conductivity.

EXPERIMENTAL

Copper films were deposited on glass substrates (20 × 20 × 1 mm cut from microscope slide) by resistive evaporation from tungsten boats at room temperature and at a base pressure of 2×10^{-6} mbar. The purity of copper was 99.98%. An Edwards (Edwards E19 A3) coating plant was used. Evaporation performed at five steps, and 5 Cu films produced at 20, 60, 110, 180 and 270 nm thicknesses. Film thickness and deposition rate were measured by a quartz crystal deposition rate controller (Sigma Instruments, SQM- 160, USA) positioned close to the substrate. Deposition angle and deposition rate were 14º and 2 Å/s for all samples. Prior to deposition, all glass substrates were ultrasonically cleaned in heated acetone then ethanol. The surface roughness of the substrates was measured by a Talysurf profilometer. The rms substrate roughness R_q was 0.3 nm. The nanostructure of these films were obtained using a Philips XRD X'pert MPD Diffractometer (CuK_α radiation) with a step size of 0.02° and count time of 1 s per step, while the surface physical morphology and roughness was obtained by means of AFM (Park Scientific) analysis. The electrical resistivity and Hall coefficient measured by four point probe instrument and a Hall Effect measurement system with a magnetic field strength of 0.25 T, respectively.

RESULTS AND DISCUSSION

X-ray Diffraction

Figure 1 shows XRD patterns of the copper thin film with different thicknesses. X-ray diffraction pattern of copper film with 20 nm thickness (sample I), showed amorphous structure. When film thickness increased to 60 nm (sample II) a weak pick is observed that can be attributed to Cu(111) crystallographic orientation (with reference to JCPDS Card No: 04-0836 and 2θº = 43.297º, system: cubic and space group: 225). Copper films of 110, 180 and 270 nm thicknesses (Sample III, IV and V, respectively) show that by increasing the film thickness the intensity of this peak is increased and one may distinguish a growth of Cu(111) preferred orientation, while a second weak peak is also appeared in the XRD pattern of the thicker films of 180 and 270 nm thickness that can be designated to Cu(220) crystallographic orientation (with reference to JCPDS Card No: 04-0836 and 2θº = 74.130º, system: cubic and space group: 225). X-ray diffraction results show that the increasing of film thickness promotes the preferred (111) orientation but not (200) orientation because Cu has the face centered cubic (FCC) crystal structure and for this structure (111) face has the lowest surface energy [15]. The growth of this preferred orientation is in agreement with the results of Chan et al. [7] who investigated copper thin films that produced by DC magnetron sputtering technique.

Crystallite size (coherently diffracting domains), D was obtained by applying the Scherrer formula (D=kλ/Bcosθ) to measure the full width at half maximum (FWHM) of the dominant peak Cu(111) in the XRD patterns of Figure 1. Where k is unity, λ = 1.54248 Å, B = FWHM that calculated from $B^2 = B_M^2 - B_s^2$ (B_M, FWHM of sample and B_S, FWHM of standard sample) and θ is the peak position in radian. The crystallite size (coherently

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diffracting domains) results are given in column 4 of Table 1. The results show that grain size increases with film thickness that is consistent with Messier et al. [16] proposed modified structural zone model (SZM). Messier et al. reported a revised SZM, using SEM, TEM and FIM, in which evolutionary growth development of physical structure, column/void sizes are assigned as five sub-zones with sizes: 1–3, 5–20, 20–40, 50–200, and 200–400 nm. This shows that each level of physical structure is about a factor of 3 larger than the previous level Further Messier [17] in an attempt to quantify morphology of thin films at low adatom mobility, proposed a fractal model that results from natural clustering during random ballistic aggregation of atoms. Savaloni et al. [18,19] also found that by increasing the film thickness the grain size increases, while its morphological and crystal structure also change.

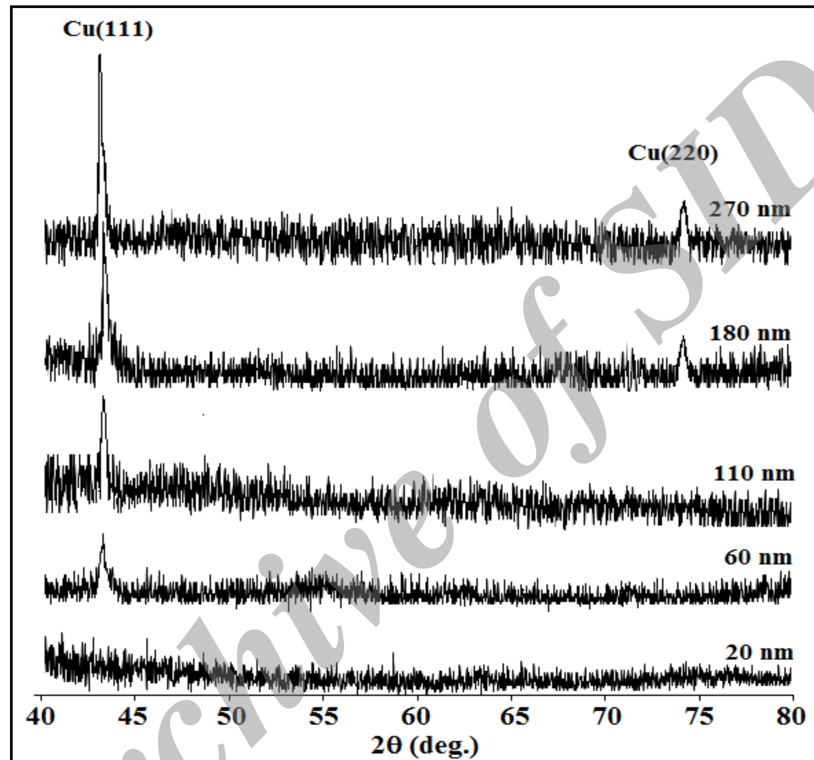

Fig. 1. XRD patterns of Copper thin films at all of thicknesses.

Table 1. Results of XRD and AFM analysis

Sample	Thickness (nm)	Grain diameter (nm) By AFM	Grain diameter (nm) by XRD	Roughness (Å) Rms	$R_{ave.}$
I	20	-	-	24.5	18.9
II	60	46.5	6.2	47.1	28.9
III	110	64.3	11.1	81.4	69.2
IV	180	85.2	13.8	93.0	73.7
V	270	90.3	18.4	93.8	74.2

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Atomic force microscopy

2D and 3D AFM images of copper thin film samples produced in this work with different thicknesses are given in Figures 2(a-e) and 3(a-e), respectively. Figures 2a and 3a show that copper film of 20 nm thickness has an amorphous structure and no visible grain or grain boundary can be observed. This is consistent with XRD result for this sample. By increasing the film thickness to 60 nm, needle like structure can be seen on the surface of the sample (Figures 2b and 3b), while at 110 nm thickness larger grains are formed (Figures 2c and 3c). At the higher thicknesses of 180 and 270 nm (Figures 2d and 2e and 3d and 3e) distinguished granular structure is formed.

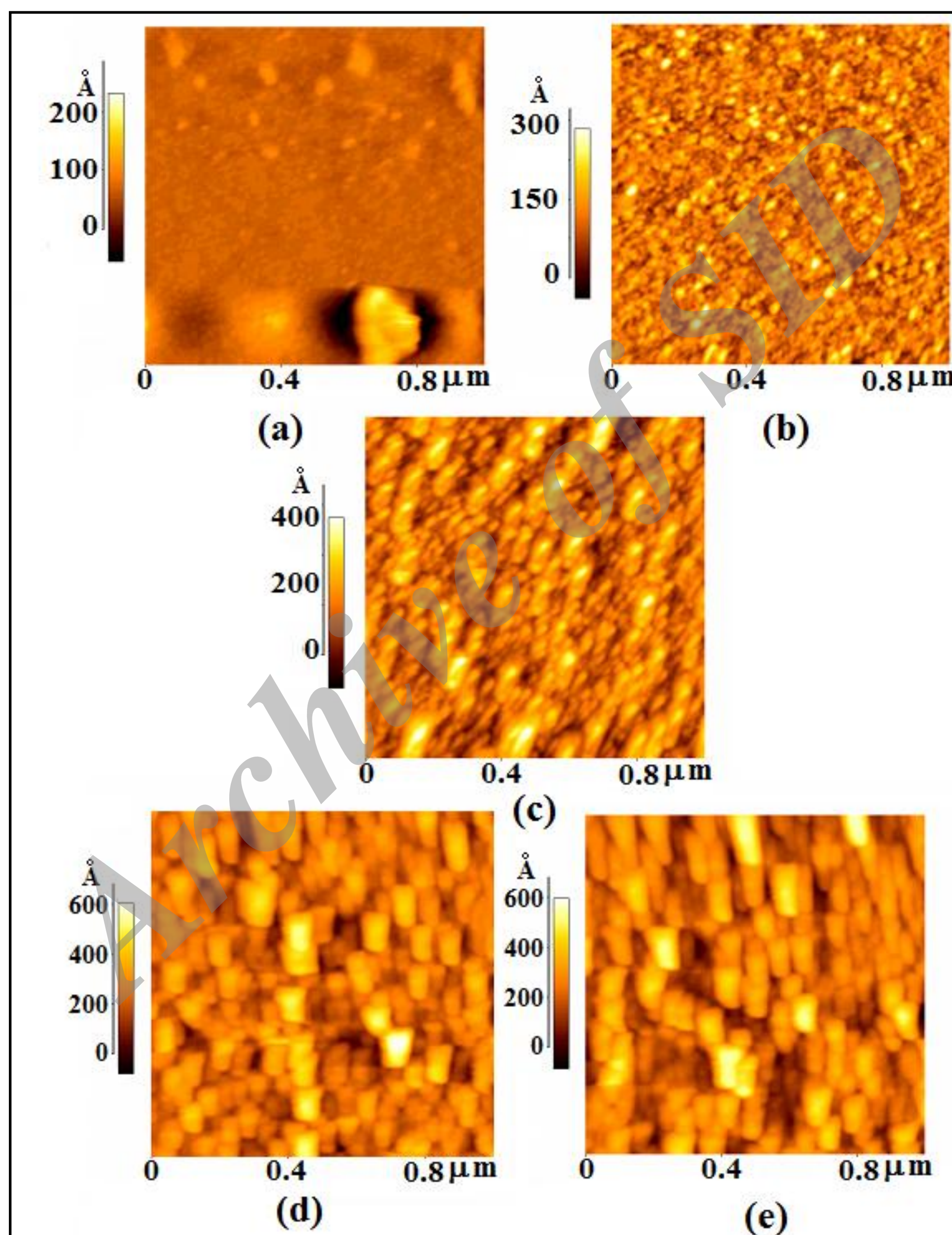

Fig. 2. 2D AFM images of Copper thin films at all of thicknesses.
a) 20 nm, b) 60 nm, c) 110 nm, d) 180 and e)270 nm.

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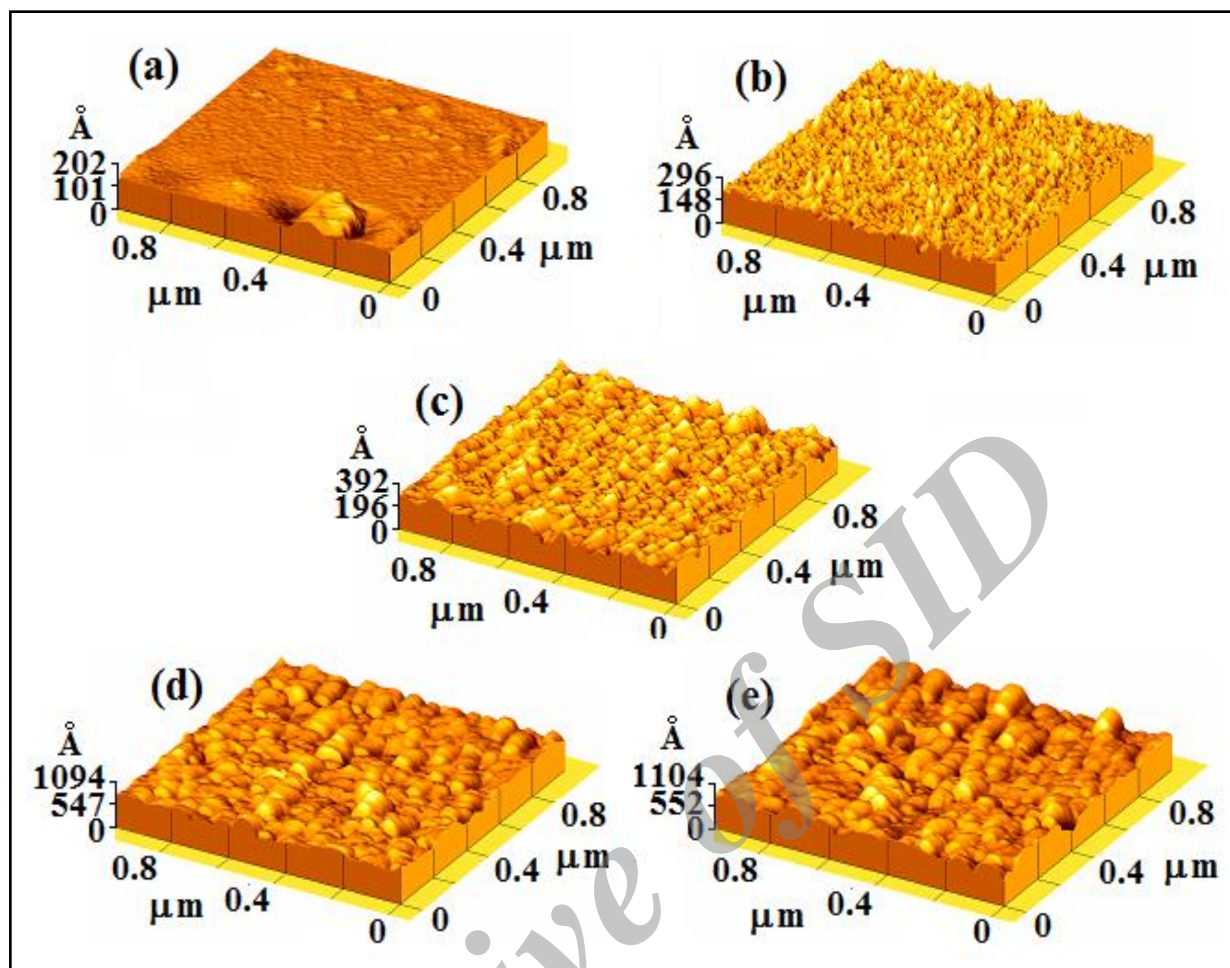

Fig. 3. 3D AFM images of Copper thin films at all of thicknesses.
a) 20 nm, b) 60 nm, c) 110 nm, d) 180 and e)270 nm.

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The sizes of the grains were calculated from 2D AFM images, using JMicrovision Code. Figure 4 shows grain size (grain diameter) distribution, while the average grain sizes of the samples are given in column 3 of Table 1.

Size distribution of grains for sample II (with 60 nm thickness) shows a narrow peak at about 40 nm. However with increasing the film thickness to 110 nm this distribution spreads out towards larger grains. With further increase of film thickness to 180 nm a broad peak is observed and its peak extends up to 100 nm. Film with 270 nm thickness shows a clear peak at 100 nm while its distribution is almost Gaussian. These results are consistent with the crystalline size obtained from the XRD analysis and agree well with the Structural zone model (SZM) predictions [16].

Root mean square (rms) and average surface roughness of copper films produced in this work obtained from AFM analysis are given in columns 5 and 6 of Table 1 and in Figure 5. There is an increase of surface roughness up to about 140-150 nm and thereafter it almost remains unchanged. The initial sharp increase can be due to mobility/surface diffusion of smaller grains on films of lower thickness that cause formation of larger grains as discussed above and can be seen in Figure 4. However when the film thickness increased to 180 and 270 nm it seems that these films have become thicker by the growth process, because the samples produced in this work were made at room temperature. Hence the last two thicknesses almost show almost the same uniform morphology.

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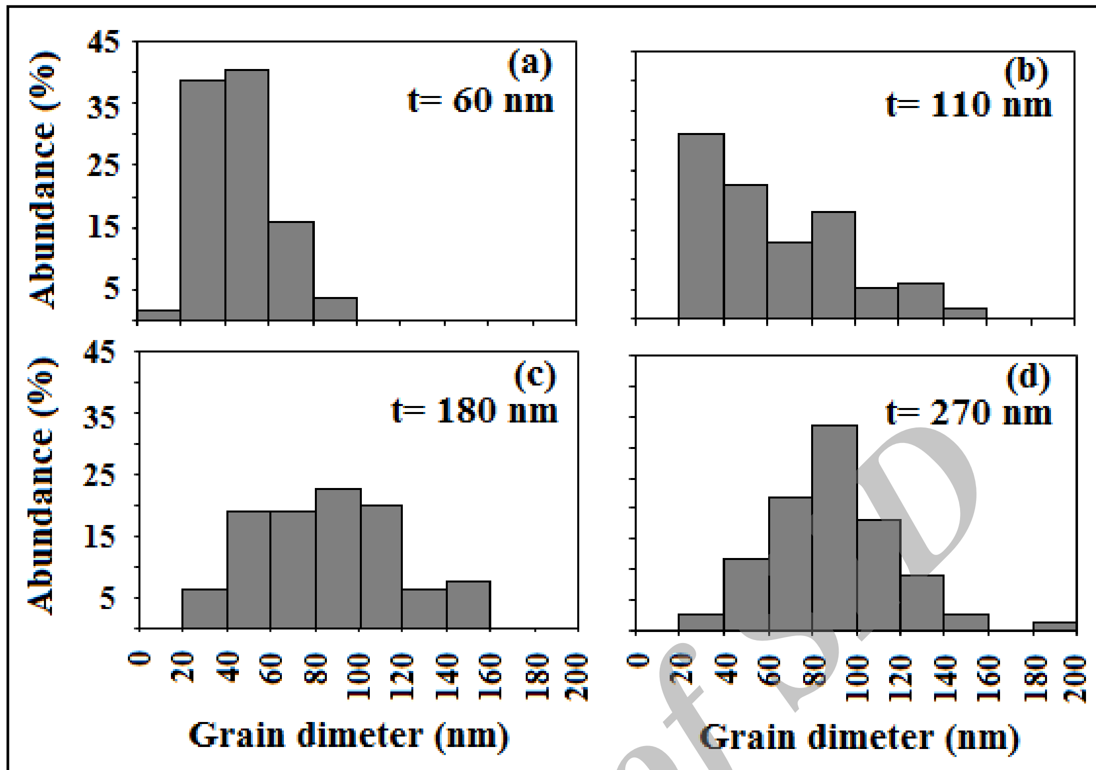

Fig. 4. Distribution functions of Grain diameter for copper thin films at thickness of, a) 60 nm, b) 110 nm, c) 180 and d)270 nm.

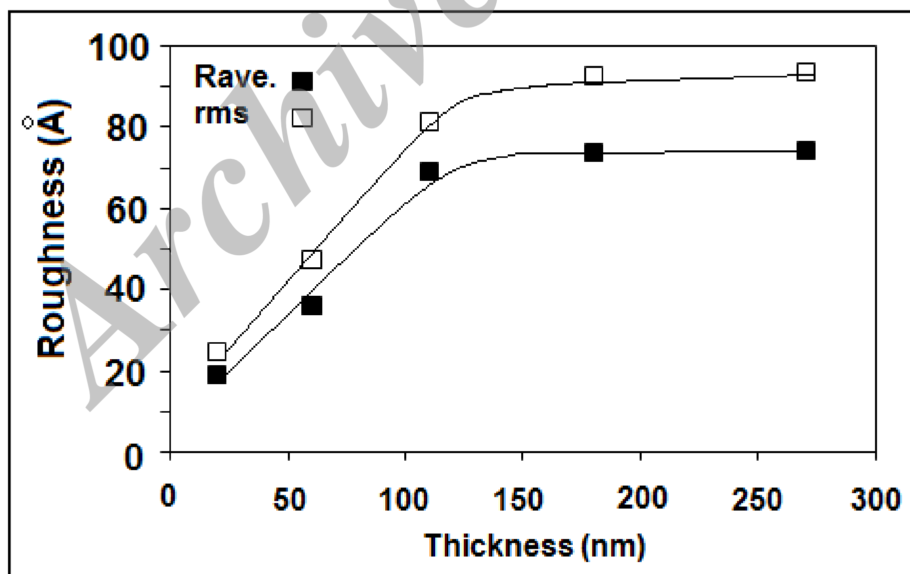

Fig. 5. Variation of rms and average roughness for copper thin films as function of film thickness.

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Four point probe

Results of dc resistivity measurement obtained using four point probe instrument are discussed in this section. In order to investigate the influence of possible (low frequency) charging effects at the electrical contacts and leads, current-voltage (I-V) curves were recorded, scanning the voltage both in increasing and decreasing increments. Figure 6 shows the results for all copper thin film samples produced in this work. The results measurements in the decreasing direction of the voltage are not shown as they matched the results in the increasing voltage direction and only clutter the figure unnecessarily. However, it should be noted that the current was kept low enough, in order not to heat up the samples. Linear I-V curves were obtained for all samples, independent of the scan direction, and there was no indication of hysteresis effect. In order to investigate the anisotropy effect in these samples, the I-V curves measurements were also carried out in four different directions on the samples, namely, two vertical (along the sample length and normal to the length) and two diagonal directions. The values of average resistivity for all samples at room temperature are given in column 3 of Table 2 and Figure 7. The results show that resistivity decreases with increasing the film thickness, which can be explained as follows:

Thin films resistivity is function of different parameters, such as surface scattering, grain boundary, impurities and different deposition parameters. Surface scattering is effective when thickness is comparable with mean free path of electrons, so that increasing of thickness decreases scattering from surface and increases conductivity. For this reason, resistivity of copper thin films decreases with increasing the film thickness. The grain boundary is effective on scattering of carriers, too. Further more, in X-ray diffraction and atomic force microscopy results it was observed that the grains size increased and the number of grain boundaries decreased with increasing the film thickness, hence they should result in decreasing of carriers scattering which in turn decreases the film resistivity.

Furthermore, the results show that the value of resistivity come to the bulk state value (1.67 μΩ.cm) at approximately 160 nm thickness that is consistent with result of Paik and Shi and et al. for copper thin films prepared by magnetron sputter type negative ion source [6] and filtered cathodic vacuum arc technique [14], respectively.

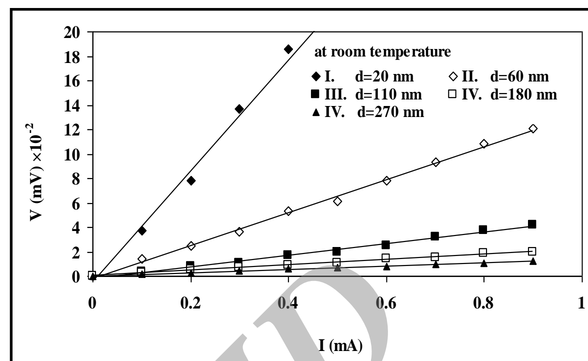

Fig. 6. Variation of voltage as a function of current for Copper thin films at all of thickness.

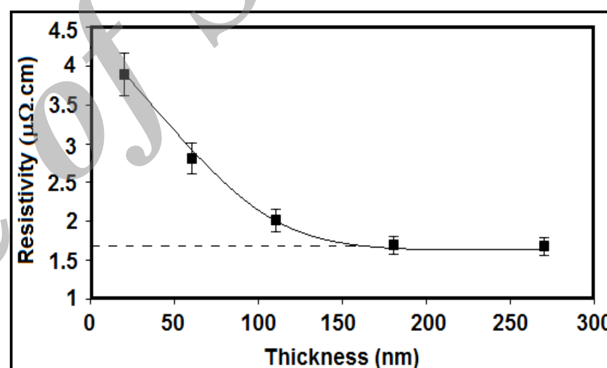

Fig. 7. Variation of resistivity for copper thin films as function of film thickness at room temperature (the dashed line is bulk state value of resistivity).

Hall effect investigation

Figure 8 shows the variation of Hall coefficient (R_H), concentration of carriers (n) and mobility (μ) as function of film thickness at room temperature, while the numerical data is presented in Table 2. The results show the increasing of thickness caused decreasing of hall coefficient and mobility and increasing of carrier concentration to about 160 nm thickness and thereafter they almost remains unchanged. So that, the value of concentration of carriers comes to bulk state value (8.47×10^{22} cm^{-3}) at about 160 nm thickness.

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Table 2. Results of Four point probe and Hall effect analysis

Sample	Thickness (nm)	Resistivity (μΩ.cm)	Hall coefficient (cm^3 A^{-1} s^{-1}) ×10^{-4}	Carrier concentration (cm^{-3}) ×10^{22}	mobility (cm^2 $V^{-1}s^{-1}$) ×10^3
I	20	3.89	2.71	2.3	6.94
II	60	2.81	1.54	4.0	4.66
III	110	2.01	0.89	7.0	4.42
IV	180	1.69	0.75	8.3	4.36
V	270	1.68	0.74	8.4	4.35

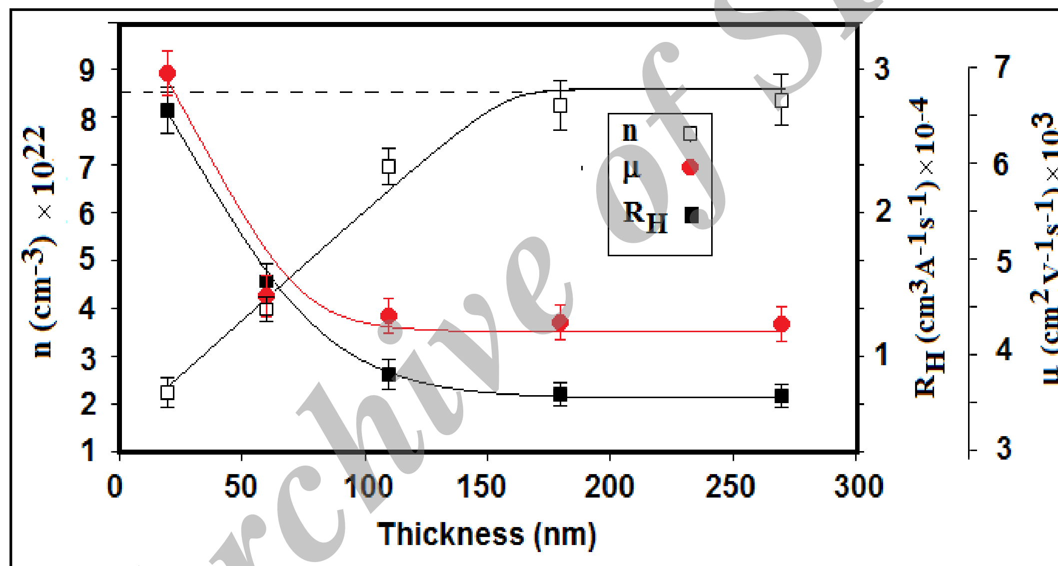

Fig. 8. Variation of Hall coefficient, carrier concentration and mobility for copper thin films as function of film thickness at room temperature (the dashed line is bulk state value of carrier concentration).

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This observation can be related to the more concentration of carriers in bulk state than thin film that caused increasing of concentration of carriers by increasing of sample thickness and is consistent with results of resistivity measurements. Increasing concentration of carriers by increasing of thickness will result in decreasing of mobility and hall coefficient as $R_H = -\frac{1}{ne}$ formula.

CONCLUSIONS

Copper thin films of different thicknesses (20-270 nm) were produced, using resistive heat deposition on glass substrates. Nano-structure of the films was obtained using X-ray diffraction (XRD), while morphology and grain size of the films were investigated by atomic force microscopy (AFM). A four point probe instrument was employed for resistivity measurement and a Hall effects measurement system was used for Hall

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effect measurement. The XRD results showed growing of Cu(111) preferred orientation by increasing the film thickness that can be related to the lowest surface energy of (111) face for (FCC) crystal structure. The grain size was calculated from both XRD and AFM analysis and the results were consistent; though with much smaller values for the XRD analysis. This may show that the films/grains are formed as single crystals. The results showed that increasing of film thickness causes increasing of grain size, roughness, concentration of carriers and decreasing of resistivity, hall coefficient and mobility.

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REFERENCES

[1] Nitta T., Ohmi T., Otsuki M., Takewaki T., Shibata T., (1992), Electrical Properties of Giant-Grain Copper Thin Films Formed by a Low Kinetic Energy Particle Process. *J. Electrochem. Soc.*, 139: 922-927.

[2] Yang C. Y., Jeng S. J.,Chen J. S., (2002), Grain growth, agglomeration and interfacial reaction of copper interconnects. *Thin solid film*. 420-421: 398-402.

[3] Patten J. W., McClanahan E.D., Johnston J. W., (1971), Room-Temperature Recrystallization in Thick Bias-Sputtered Copper Deposits. *J. Appl. Phys.* 42: 4371-4378.

[4] Liu C. S., Chen L. J., (1996), Effects of substrate cleaning and film thickness on the epitaxial growth of ultrahigh vacuum deposited Cu thin films on (001)Si. *Appl. Surf. Sci.*, 92: 84-88.

[5] Nakai H., Fudaba K., Shinzawa K., Hashimoto M., (1998), Electrical property on copper thin film with chromium under-layer. *Thin solid films.* 317: 202-205.

[6] Paik N., (2005), Characteristics of Cu films prepared using a magnetron sputter type negative ion source (MSNIS). *Nuclear Instruments and Methods in Physics research B.* 229: 436-442.

[7] Chan K. Y., Tou T. Y., Teo B. S., (2006), Thickness dependence of the structural and electrical properties of copper films deposited by dc magnetron sputtering technique. *Microelec. J*. 37: 608-612.

[8] Qiu H., Wang F., Wu P., Pan L., Tian Y., (2002), Structural and electrical properties of Cu films deposited on glass by DC magnetron sputtering. *Vac.* 66: 447-452.

[9] Fang T. H., Chang W. J., (2003), N anomechanical properties of copper thin films on different substrates using the nanoindentation technique. *Microelec. Eng.*, 65:231-238.

[10] Savaloni H., Khakpur M. R., (2003), Optical properties of thin film Cu films as function of substrate temperature. *J. Sci.* 14(1): 71-79.

[11] Suresh S., Nieh T. G. and Choi B.W., (1999), Nano-insentation of copper thin films on silicon substrate. *Choi, Scr. Mater.* 41(9): 951-957.

[12] Boo J. H., Jung M. J., Park H. K., Nam K. H., Han J. G., (2004), High-rate deposition of copper thin films using newly designed high-power magnetron sputtering source. *Sur. Coat. Tech.*, 188–189: 721-727.

[13] Cho N. I., Nam H. G., Choi Y., Yang J. S., (2003), C hemical vapor deposition of copper thin films for multi-level interconnections. *Microelec. Eng.* 66: 415-421.

[14] J. R. Shi, S. P. Lau, Z. Sun, X. Shi, B. K. Tay, H. S. Tan, (2001), Structural and electrical properties of copper thin films prepared by filtered cathodic vacuum arc technique. *Sur. Coat. Tech.* 138: 250-255.

[15] N. Joshi, A.K. Debnath, D.K. Aswal, K.P. Muthe, M. Senthil Kumar, S.K. Gupta, J.V. Yakhmi, (2005), Morphology and resistivity of Al thin films grown on Si (1 1 1) by molecular beam epitaxy . *Vac.* 79: 178-185.

[16] Messier R., Giri A.P., Roy R.A., (1984), Revised structure zone model for thin film physical structure. *J. Vac. Sci. Technol. A.* 2: 500-503.

[17] Messier R., (1986), Toward quantification of thin film morphology. *J. Vac. Sci.Technol. A*, 4: 490-495.

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[18] Savaloni H., Player M. A., (1995), Influence of deposition conditions and substrate on the structure of UHV deposited erbium film. *Vac*. 46: 167-.179.

[19] Savaloni H., Player M. A., (1995), Morphological changes in UHV deposited Er/a C films: nucleation, growth and grain structur. *Thin Solid Films*. 256: 48-58.

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