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SEM AND XRD CHARACTERIZATIONS OF NANOGRANULAR COPPER METAL FILMS

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Key words: Copper; Sputtering; Film thickness; Structural properties; Morphological properties

Abstract: This paper reports on the influence of film thickness on the morphological and structural properties of direct current (DC) magnetron sputter-deposited nanogranular copper (Cu) metal films. Cu metal films with thicknesses of 130 nm to 1800 nm were deposited on silicon substrates at sputtering power of 125 W in argon working gas pressure of 3.6 mTorr at room temperature. The morphological and structural properties of the nanogranular Cu metal films were investigated by scanning electron microscopy (SEM) and X-ray diffractometer (XRD). Results from our experiment show that the Cu nanograins grow with increasing film thickness, along with enhanced film crystallinity. Possible mechanisms of film thickness dependent microstructure formation of the nanogranular Cu metal films are discussed in the paper, which explain the interrelationship of grain growth with increasing Cu metal film thickness.

SEM and XRD Characterizations of Nanogranular Copper Metal Films

Kjučne besede: baker, nanašanje, debelina filma, strukturne značilnosti, morfološke značilnosti

Izvleček: Članek poroča o vplivu debeline filma na morfološke in strukturne značilnosti z DC magnetronom nanešenega nanozrnatega bakrenega filma. Bakreni kovinski filmi z debelino od 130nm do 1800nm so bili nanešeni na silicijeve podlage z močjo 125 W v atmosferi argona pri pritisku 3.6m tora pri sobni temperaturi. Morfološke in strukturne značilnosti nanozrnatih bakrenih kovinskih filmov smo pregledovali z vrstično elektronsko mikroskopijo (SEM) in difraktometrom za X-žarke (XRD). Rezultati naših poskusov kažejo, da bakrena nanozrna rastejo s povečevanjem debeline skupaj s povečano kristalnostjo filma. V članku obravnavamo tudi možne mehanizme, ki povzročajo odvisnost mikrostrukture nanozrnatih bakrenih filmov od njegove debeline.

1. Introduction

Copper (Cu) metal thin films with nano geometric and microstructural dimensions are ubiquitous of modern technology, with applications spanning the range from the catalysis to microelectronic devices. In ultralarge scale integrated (ULSI) circuits, achievement of maximum signal transmission with minimum signal propagation delay along metal interconnects in emerging chip and system architectures requires minimizing the resistance-capacitance (RC) time delay. Minimizing RC delay in thinner interconnects has forced a transition from aluminum (Al) based interconnects to Cu metallization schemes owing to the higher conductance of Cu, which not only reduces the delay time but also enables higher current densities at lower voltages, minimizing heat generation and power requirements and increasing the packaging densities per metallization level. Apart from reduction in RC time delay, Cu is more promising than Al for metallization in terms of electromigration (transport of atoms of the metal interconnects subjected to high current densities) resistance and mechanical elasticity stress /1/. Cu exhibits superior resistance to electromigration and stress-induced voiding open-circuit failure as its atoms are more strongly bounded together which account for less likely to fracture under stress comparing to Al.

Due to the noticeable advantages as interconnection candidate in terms of low electrical resistivity, good electromigration resistance and high melting temperature /2/, the studies of Cu thin films have been generally centered on the electrical and microstructural properties of the Cu films deposited with various deposition techniques. Among these, magnetron sputter deposition is a well established technology and offers an attractive alternative to diode techniques, due to lower working gas pressure employed which makes possible higher sputtering rate /3/. Also, magnetron sputtering is an attractive mass production technique due to the possibility of large area deposition /4/, which makes it an effective economic approach in microelectronics manufacturing.

In our previous research work, we studied the influence of sputtering power, deposition pressure and substrate temperature on the material properties of direct current (DC) magnetron sputter-deposited Cu metal films /5,6/. We have also examined the influence of Cu film thickness using atomic force microscopy (AFM) /7/. In this paper we present the investigation results on the effect of film thickness on the morphological and structural properties of nanogranular Cu metal films using scanning electron microscopy (SEM) and X-ray diffractometer (XRD).

2. Experimental

All Cu films were deposited with a DC magnetron sputtering system (Fig. 1) using a circular 5.08 cm diameter Cu target of 99.995 % purity. The target to substrate distance was 9 cm. The substrates for the deposition were 6 mm x 12 mm p-type silicon (Si) and were cleaned follow standard cleaning procedure. The base pressure was lower than 2 x 10^{-6} Torr, achieved with a cryo pump coupled with a rotary pump. The sputtering deposition was carried out at a working pressure of 3.6 m Torr in high purity argon (Ar) gas (99.999%) with DC sputtering power of 125 W, which led to a deposition rate of approximately 50 nm/min. In order to study the influences of the film thickness, a series of Cu metal film samples with thicknesses controlled at around 130 nm to 1800 nm were fabricated at room temperature. The thicknesses of these Cu films were checked in situ with a quartz crystal monitor located near the substrate during the sputtering process, and was quantified with the Mahr surface profilometer after the deposition process by measuring the step height between masked and unmasked regions on the substrate. The surface morphology and microstructure of the Cu metal films were examined by means of SEM. XRD analyses in the θ-2θ mode using Cu Kα X-rays were carried out to investigate the crystallinity and orientation of the Cu metal films with different thicknesses. The deposition conditions of the Cu metal films presented in this work are summarized in Table 1.

Table 1. Deposition parameters for the Cu metal films.

Target	Cu
Substrate	p-type Si
Target-substrate distance	9 cm
Ar working gas pressure	3.6 m Torr
Sputtering power	125 W
Deposition rate	~50 nm/min
Film thickness	130 nm - 1800 nm

3. Results and discussion

The influence of the thickness on the surface morphology of the nanogranular Cu metal films was examined by the SEM, as shown in Fig. 2a-d. The SEM images show the features of structural change and grain growth for nanogranular Cu metal films with different thicknesses ranging from 130 nm to 1800 nm. The SEM image of the 130 nm thick Cu metal film deposited at room temperature (Fig. 2a) exhibits uniform surface, indicating its amorphous-like behavior /8/. Fig. 2b, 2c, and 2d show the SEM images for Cu metal films with thicknesses of 275 nm, 500 nm and 1800 nm, respectively. In these images, nanograin features develop in the Cu metal films and are enhanced with increasing film thickness. The nanograins are smaller and less definable producing smoother surface in image of Fig. 2b, and larger and evenly scattered in Fig. 2c. In Fig. 2d, the grains were observed to have agglomerated together and formed smaller number of bigger grains. The nanograins are homogeneously distributed all over the substrate surfaces. Comparing the SEM images in Fig. 2, Cu metal film with higher thickness clearly exhibits profound grain features. This can be explained by the film structural evolution with thickness, in which the grains grow with increasing film thickness /9/. The transformation from the uniform smooth amorphous-like film structure to the fine nanogranular features, followed by the growth in the nanograins for the Cu metal films with increasing film thickness as demonstrated in Fig. 2 can be attributed to the surface energy minimization during the growth process to achieve thermodynamical equilibrium. As the growth proceeds, those grains with preferred growth direction survive due to surface tends to evolve towards a situation of low surface energy. This results in evolution of large grained columnar morphology from a much larger number of fine grains which were originally nucleated on the substrate / 10/, and improved film texture according to following equations /11/:

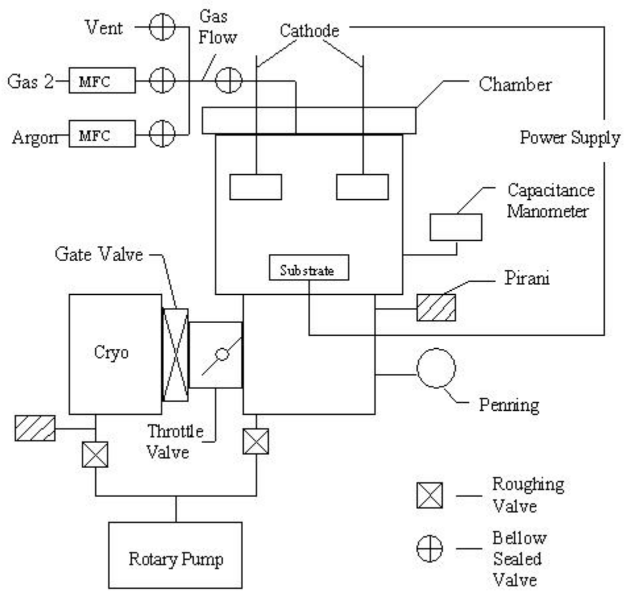

Fig. 1: Schematic diagram of the magnetron sputtering deposition system.

$$\frac{dr_i}{dt} = K[\gamma(t) - \gamma_i], \qquad (1)$$

where r_i, g_i and $g(t)$ are the radius, the surface energy of i grain and mean surface energy of the surrounding grain of i. K can be described as:

$$K = (\frac{v_o L}{kT}) V_{at}^{\frac{2}{3}} \exp(\frac{-E}{kT}), \qquad (2)$$

where v_o, L, k, T, $V_{at}^{2/3}$ and E are the attempt frequency, the thickness of a small layer near the surface, Boltzmann factor, the temperature, the surface change in each grain for a single jump and the mean activation energy.

The improved film texture is in agreement with the XRD patterns for these nanogranular Cu metal films with different thicknesses, as shown in Fig. 3. The intensity of a peak in X-ray diffraction pattern is a direct measure of the film crystallinity. From the intensity of the diffraction peaks (Fig. 4), Cu metal film with higher thickness exhibits enhanced

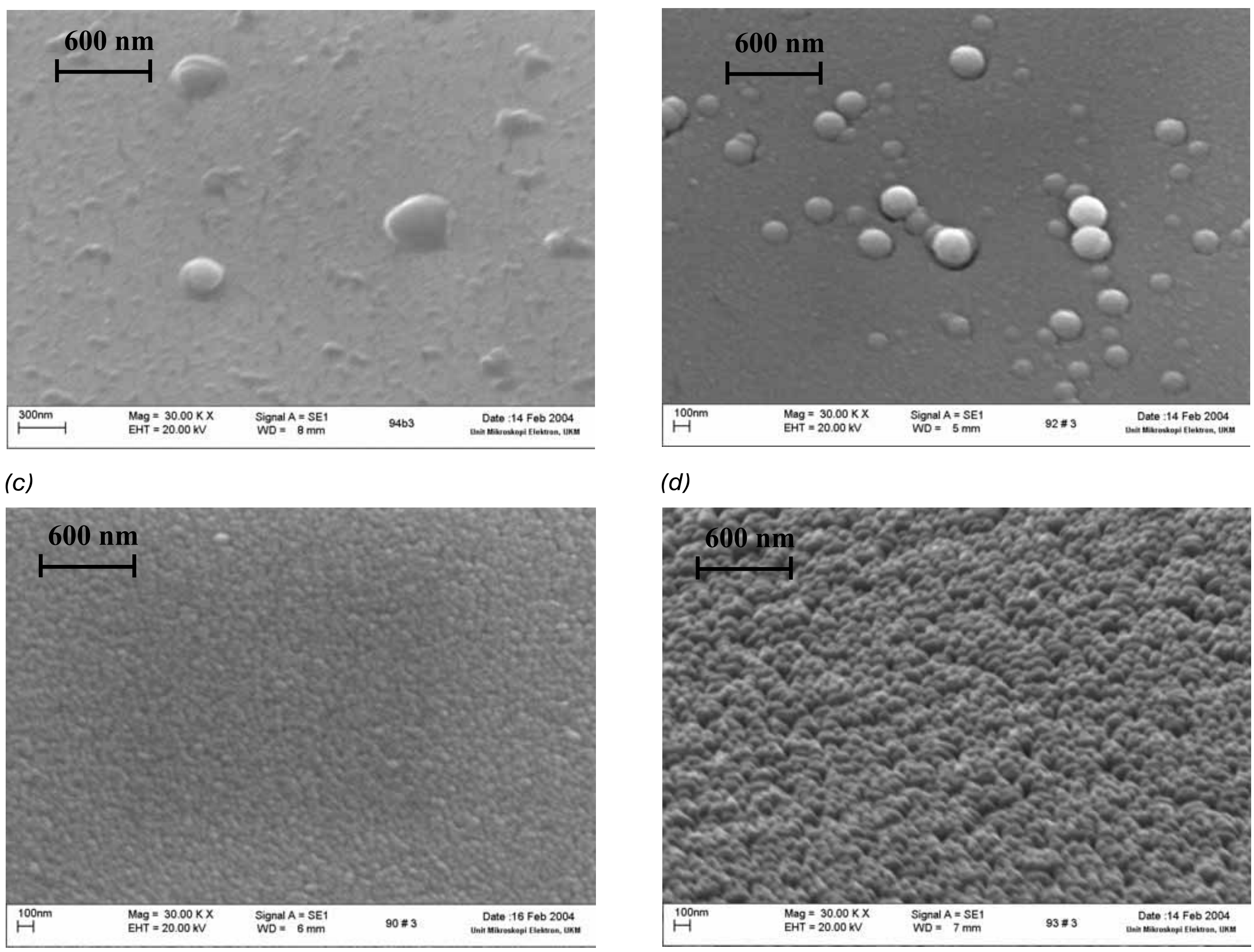

Fig. 2: Scanning electron microscopy (SEM) images of nanogranular copper (Cu) metal films with thicknesses of (a) 130 nm, (b) 275 nm, (c) 500 nm, and (d) 1800 nm.

crystalline nature than thinner metal film with preferential orientation of (111) observed at 2θ = 43 °. The film thickness has no significant influence on the preferred (200) orientation for the DC magnetron sputter-deposited Cu metal films. The increasing film thickness promotes the

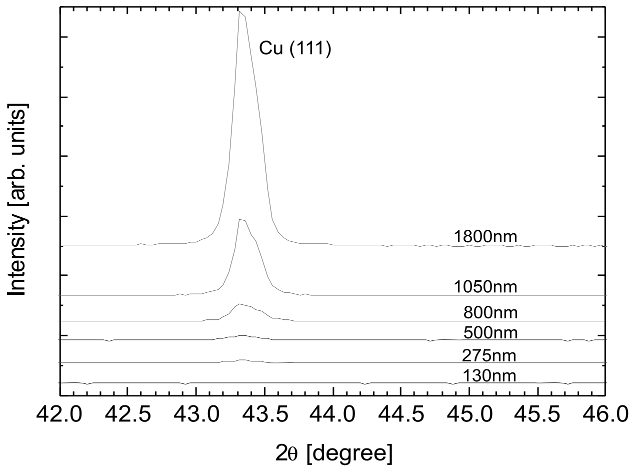

Fig. 3: X-ray diffraction (XRD) patterns of the copper (Cu) metal films with different thicknesses.

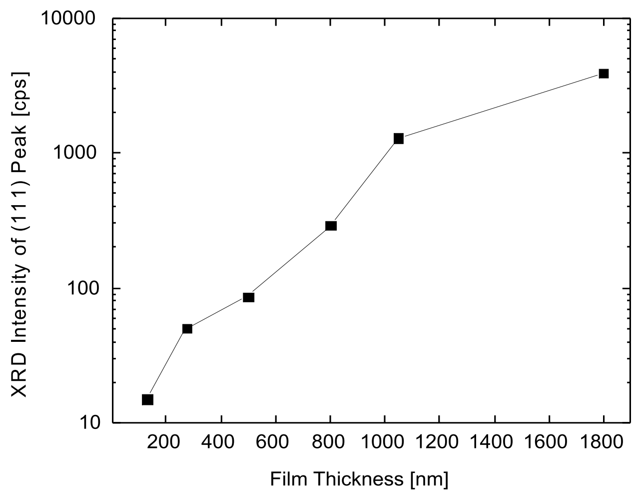

Fig. 4: Intensity of X-ray diffraction (XRD) (111) peaks of the copper (Cu) metal films as a function of the film thickness.

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 /12/.

In order to obtain deeper insight into the influence of film thickness on the microstructure, we calculated the grain size of the Cu metal films with different thicknesses. Grains or crystallites are crystal units in a material that diffract in phase. When related to metallic films, the term 'grain' is also used to represent the crystal units in metallic films. In principle, the analysis of the grain size in XRD diffraction patterns is performed by measuring the broadening of a particular peak in a diffraction pattern associated with a particular planar reflection from within the crystal unit cell. The grain size (G) of the Cu metal films was deduced from the (111) diffraction peak using Scherrer's formula:

$$G = \frac{K \cdot \lambda}{FWHM \cdot \cos\theta}, \tag{3}$$

where K is the shape factor of the average grain (0.9), λ is the X-ray wavelength (0.154056 nm for Cu $K\alpha_1$), FWHM is the full width at half maximum of the diffraction peak, θ is the diffraction peak angle. From the Eqn. (3), the grain size is inversely related to the FWHM of the diffraction peak. The more narrow the diffraction peak, the larger the grain size is. This is due to the periodicity of the individual grain domains which are in phase, reinforcing the X-ray beam, and results in a narrow diffraction peak. Most thin films have some degree of preferred orientation. For Cu, it is usually (111) crystallographic direction that has the greatest degree of preferred orientation. Therefore, the measured grain size of the (111) diffraction peak represents the grain size in the columnar direction, normal to the surface of the substrate. Fig. 5 demonstrates the dependence of the Cu metal grain size and FWHM on the film thickness. In general, the grain size increases with increasing Cu metal film thickness and the FWHM acts in opposite way. For the Cu metal films with thicknesses of 130 nm, 275 nm, 500 nm, 800 nm, 1050 nm, and 1800 nm, the corresponding nanograin sizes are 27 nm, 36 nm, 39 nm, 44 nm, 48 nm, and 51 nm, respectively, while the corresponding FWHM are 0.40 °, 0.30 °, 0.28 °, 0.25 °, 0.22 °, and 0.21 °, respectively. The grain size of the Cu metal films deduced using Scherrer's formula is consistent with the intensity of the corresponding diffraction peak of the films.

4. Conclusions

We have qualitatively evaluated the thickness dependence of the morphological and structural properties of DC magnetron sputter-deposited nanogranular Cu metal films. The surface morphological study with SEM shows the enhanced microstructure, while the XRD patterns reveal the improved crystalline quality for the thicker Cu metal films. The grain size of the Cu metal films was found to increase with increasing film thickness, which is due to the surface energy minimization when the film gets thicker.

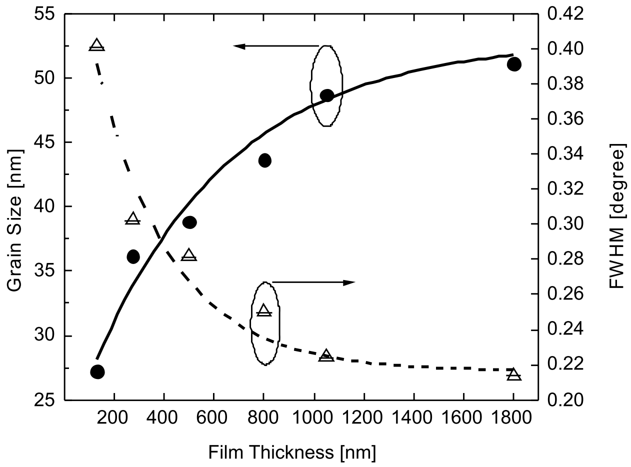

Fig. 5: Grain size and full width at half maximum (FWHM) of X-ray diffraction (XRD) (111) peaks of the copper (Cu) metal films as a function of the film thickness.

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