Effect of Sputtering Power on the Nucleation and Growth of Cu Films Deposited by Magnetron Sputtering

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Cu thin films were deposited on Si(100) substrates using direct current (DC) magnetron sputtering. We focused on the effect of sputtering DC power on the electrical, structural properties, and the nucleation and growth of Cu films during the initial stage of sputtering. The Cu films deposited at higher sputtering power showed strong crystallinity, low electrical resistivity in comparison with the Cu films deposited at lower sputtering power. Concerning the nucleation and growth of Cu films during the initial stage of magnetron sputtering, it was found that the progress of the nucleation and growth of the Cu films at higher sputtering is much faster than those of the Cu films at lower sputtering power even though they have a similar nucleation and growth mechanism, and their relation to resultant microstructure was confirmed by atomic force microscopy.

Keywords: copper, thin films, magnetron sputtering, atomic force microscopy, nucleation, growth

1. Introduction

Copper is an attractive interconnecting material for current Si ultra-large scale integrated (ULSI) devices due to its low resistivity and super resistance to electro-migration. The electrical resistivity of Cu films is an important factor for using interconnects.¹⁻³ Thin films of Cu are deposited by various techniques such as evaporation, sputtering, chemical vapor deposition, electroplating, and ion beam deposition.⁴⁻¹¹ Among these techniques, direct current (DC) magnetron sputtering is one of the best methods for practical preparation of Cu films. Since DC magnetron sputtering technique has the advantage of much better productivity than other deposition methods, it is widely used as mass production processes.

Generally, structure and electrical qualities of films strongly depend on the deposition process. The behavior of electrical property of Cu films complies with direct current (DC) power and deposition time. DC power variation is an important factor to essentially control properties of Cu films. Higher DC power helps to increase the surface mobility and this results in forming a large crystallite size which may be due to high adatom energy, and thereby highly crystalline Cu films can be formed.⁵⁻⁷ Therefore, it can be anticipated that the changes in DC power in the sputtering system plays an important role in the nucleation and growth of Cu films as well as electrical and structural properties. However, most previous works have not focused on the nucleation and growth of Cu films at different sputtering power during the initial stage of deposition. Considering the above factors, we focused on the nucleation and growth of Cu films deposited by DC magnetron sputtering using atomic force microscope (AFM) as a function of different sputtering powers and deposition times.

2. Experimental Procedure

Substrates used in the present experiment were p-type (100) oriented Si wafers with a resistivity of 0.03⁻0.25Ω·m, and thickness of ~725µm, cut into 10 × 20 mm pieces. The Si substrates were cleaned in diluted HF solution (5%) for 1 min, and were rinsed in de-ionized water prior to loading into a vacuum chamber. The sputtering chamber was evacuated to 3 × 10⁻⁴ Pa base pressure using diffusion and mechanical booster pump combination prior to deposition. Before the deposition of Cu films, a Cu target (99.99% pure and 3 inch diameter) was pre-sputtered in pure argon atmosphere for 10 min in order to remove oxide layer on the surface of the target. Cu films were deposited by DC sputtering system in pure argon gas (99.999%) with pressure of 6 × 10⁻² Pa. The distance from the Cu target to the Si substrate in the experimental was 80 mm. Deposition time was 10 min while the sputtering power was varied from 0.55 to 2.74 W·cm⁻². Substrate temperature was kept constant at 25°C. The crystalline structure of the Cu films was identified by X-ray diffraction (XRD, Rigaku, Rotaflex Ru-200B). The thickness of the Cu films was measured by Mahr surface profilometer after the deposition process by measuring a step height between masked and unmasked regions on the substrate. Resistivity measurements of the Cu films were done by the four-point probe method using Advanced Instrument Technology (AIT, CTM-SR100MP) at room temperature. The four point probe head (JANDEL ENG.) was a linear type with 1 mm spacing between each probe. Load for the measurement was 0.1 kg per needle. Needles were solid tungsten carbides with φ 0.4 mm and 45° included angle. The nucleation and growth as well as the surface morphology of the Cu films were examined by atomic force microscopy (AFM, Surface Imaging Systems NanoStation-II).

3. Results and Discussion

Figure 1 shows XRD patterns of the Cu films deposited at various sputtering powers. No clear peaks were observed at 0.55 W·cm⁻². As the sputtering power increased from 1.1 to 2.74 W·cm⁻², XRD patterns clearly exhibited (111) and (200) peaks of Cu structure. This refers that the crystallinity
of the Cu film deposited at higher sputtering power becomes stronger compared to that of the Cu films deposited at lower sputtering power. Therefore, the diffraction patterns demonstrate that the DC power promotes the film crystallinity. Chan et al.\(^5\) attributed this to the mobility improvement of adatoms sputtered on the surface, which is required to form highly crystalline films. Because it is believed that high DC sputtering power in magnetron sputtering system energizes inert argon gas to provide sufficient kinetic energy to adatoms. The surface diffusion of these adatoms is then expected to enhance with the momentum transfer to the nucleation and growth of the Cu films.\(^5\)

The crystallite size of Cu films \((t)\) was evaluated from the full width half at maximum (FWHM) intensity of X-ray diffraction peak of Cu(111) using Sherrer’s relation.\(^12\)

\[
t = \frac{k \lambda}{\beta \cos \theta}
\]

where \(k\) is a constant with value about 0.9 for Cu target and \(\beta\) means the full width at half maximum (FWHM) intensity of the peak measured in radians. \(\lambda\) is the X-ray wavelength (0.154178 nm for Cu \(K\alpha_1\)) and \(\theta\) indicates diffraction peak angle. Figure 2 shows the crystallite size and thicknesses of the Cu films deposited at various sputtering powers. The thickness of the Cu films increased linearly with the sputtering power. In addition, the crystallite size of the Cu films increased from 30.9 to 69.3 nm with the increase of the sputtering power from 0.55 W/cm\(^2\) to 2.74 W/cm\(^2\). The crystallite size increased with the sputtering power indicates that the higher surface mobility caused by high adatom energy strengthens the crystallinity of the Cu films at higher sputtering power as demonstrated in Fig. 1.

Figure 3 shows the effect of the sputtering DC power on the electrical property of the Cu films. The Cu films deposited at 0.55 W/cm\(^2\) showed a high resistivity value (about \(6.00 \times 10^{-2} \mu\Omega\cdot m\)). However, when the sputtering power was increased, the resistivity of the Cu films markedly decreased, and the resistivity was about \(1.86 \times 10^{-2} \mu\Omega\cdot m\) at the high DC power of 2.74 W/cm\(^2\), which is close to the bulk value (\(1.67 \times 10^{-2} \mu\Omega\cdot m\)). Possible factors which were contributed to the considerable decrease of the Cu film resistivity at high sputtering power are considered to be an improvement of the film density as well as the large crystallite size caused by the high adatom energy.\(^5\)

Concerning the size effect of Cu films, in the present work, since the minimum of the Cu film thickness was 110 nm, which is clearly larger than the bulk electron mean free path of Cu (39 nm). On the evaluation of the resistivity, the size effect does not work on the resistivity so much when the thickness is above 100 nm even though the grain size is increasing with the film thickness.\(^10,13\) Therefore, it is believed that the increased thickness of Cu film does not clearly affect the resistivity of the Cu film in this study. On the other hand, the high value of the Cu film resistivity at lower sputtering power is attributed to the structural defects such as low density and crystallinity due to the low surface mobility. This is already confirmed from the XRD patterns and corresponding crystallite sizes deduced using Sherrer’s formula (Fig. 2).

AFM images of the Cu films deposited at various sputtering powers are shown in Fig. 4. The images of the Cu films were acquired in a 3 \(\mu\mathrm{m} \times 3\ \mu\mathrm{m}\) area. The AFM micrographs show the surface morphologies and grain growth for the Cu films deposited at 0.55, 1.10, 1.65, 2.19 and 2.74 W cm\(^{-2}\) at the deposition time of 10 min. In the case of the lowest sputtering power, the morphology of the Cu film was observed to have a uniform layer. This indicates that there are a few grains with an oriented texture, which results in the structure with a random orientation (Fig. 4(a)).
Figures 4(b)–(e) show the deposition of the Cu films at 1.10, 1.65, 2.19 and 2.74 W cm$^{-2}$ respectively. The surface morphologies showed the erection of grain feature from the film surface and this phenomenon was found to have higher intense with increasing the sputtering power. The Cu film deposited at higher sputtering power exhibits profound large grains with orientations. These morphologies are due to the fact that sputtering power helps to increase the surface mobility of adatoms, which is required to form continuous films. The surface diffusion of these adatoms was then enhanced by the higher sputtering power, which results in a provision of the momentum transfer to the growing surface.

Figure 5 demonstrates the qualitative dependence of RMS roughness of the Cu films as a function of the sputtering power. It was observed that the RMS roughness increased with increasing the sputtering power. For the Cu films deposited at various sputtering powers of 0.55, 1.10, 1.65, 2.19 and 2.74 W cm$^{-2}$, the corresponding RMS roughness are 1.81, 3.58, 4.37, 9.38 and 11.0 nm, respectively. It can be expected that low sputtering power exhibits low deposition rate which is due to less energetic argon over the target species. This affected slow nucleation and growth related to a few small grains on the surface in the present study. On the other hand, as the higher sputtering power increased, the RMS roughness increased due to the large grains as observed in the AFM micrograph in Fig. 4. This trend of RMS roughness is associated with the crystallite size (Fig. 2).

In order to investigate the difference of the nucleation and growth between the Cu films deposited at the sputtering powers of 0.55 W cm$^{-2}$ and 2.74 W cm$^{-2}$, we have focused on the initial stage of the Cu films as a function of deposition time. Figure 6 shows the 3D AFM images of the Cu films at 0.55 W cm$^{-2}$ and 2.74 W cm$^{-2}$ for 2, 5, 10 s, respectively. In order to explain distinctly the grain growth mode from 2 s to 10 s, we selected magnified 3D images in the case of 10 s deposition time. The AFM images revealed that there was a similar nucleation and growth mechanism between the Cu films at 0.55 W cm$^{-2}$ and 2.74 W cm$^{-2}$, while there existed a
time-lag of the nucleation and growth between them. In the initial nucleation stage, the main difference is only the number of nuclei on the substrate for the Cu film at lower DC power ($0.55 \text{ W/cm}^2$) compared to those on the substrate for the Cu film at higher DC power ($2.74 \text{ W/cm}^2$). Several nuclei were observed just after the deposition time of 2 s for the Cu film at the sputtering power of $0.55 \text{ W/cm}^2$. However, in the case of the sputtering power at $2.74 \text{ W/cm}^2$, a number of nuclei was observed after the deposition time of 2 s. Concerning the grain formation and growth after the deposition time of 5 and 10 s, a few Cu grains are formed and growth through coalescence of the nuclei for the Cu films at $0.55 \text{ W/cm}^2$. However, in the case of the sputtering power at $2.74 \text{ W/cm}^2$, a number of nuclei was observed after the deposition time of 2 s. Concerning the grain formation and growth after the deposition time of 5 and 10 s, a few Cu grains are formed and growth through coalescence of the nuclei for the Cu films at $0.55 \text{ W/cm}^2$ as seen in Fig. 6(c). It is considered that a typical island growth of the Cu grains was about to start from the magnified 3D image of 10 s. On the other hand, at the higher sputtering power, a typical island growth of Cu grains formed at the initial stage was considerably progressed at the deposition of 10 s by the migration of adatoms on the substrate. Therefore, it was found that the progress of the nucleation and growth of the Cu films at $2.74 \text{ W/cm}^2$ is much faster than those of the Cu films at $0.55 \text{ W/cm}^2$ due to the difference in the sputtering power even though they have a similar nucleation and growth mechanism, which can be confirmed from the magnified 3D images after the deposition time of 10 s as shown in Figs. 6(e) and (f).

There are three basic modes of film growth based on the principal interactions between substrate atoms and deposited atoms. First is (i) Volmer-Weber’s 3D island growth. Second is (ii) Frank-van de Merwe’s layer growth (2D growth). And the last is (iii) Stranski-Krastanov’s 3D clusters on top of one or several monolayers. We have reported the effect of a negative substrate bias voltage on the nucleation and growth of Cu films during the initial stage of ion beam deposition. The Cu films with or without the negative substrate bias voltage were found to have a different dependence of nucleation and growth. By applying the negative substrate bias voltage, the nucleation and growth of Cu films was changed from 3D island growth to the lateral growth by a sufficient migration of adatoms. On the other hand, in the present study, we found that the growth mode of both the Cu films at $0.55 \text{ W/cm}^2$ and $2.74 \text{ W/cm}^2$ corresponds to the typical 3D island growth, which results in a columnar structure with large grains due to an insufficient migration of adatoms on the surface as usually observed during general sputtering method. It is well known that the kinetic energy of atoms toward the substrate has an influence on the nucleation rate and nuclei density increase. In the previous study, we could confirm that the bombardment with high energy particles accelerated by applying the negative substrate bias voltage could change to a progressive nucleation and lateral growth by the sufficient migration of adatoms. On the other hand, in the present results of AFM images, we could find high sputtering DC power to progress the nucleation and the 3D island growth of Cu films without

![Fig. 6 3D AFM images of the Cu films at the sputtering power of 0.55 and 2.74 W cm\(^{-2}\) with various deposition time; (a) and (b) 2 s, (c) and (d) 5 s, (e) and (f) 10 s.](image-url)
any mechanism change. From the above experimental results, it was found that sputtering DC power has a great influence on the properties of Cu films deposited by magnetron sputtering and then the sputtering DC power is one of the important parameters to control the nucleation and growth during the initial stage of sputtering as well as the electrical and structural properties of Cu films.

4. Conclusions

Cu films were deposited on Si (100) substrates at various sputtering powers in the range from 0.55 to 2.74 W cm$^{-2}$ using DC magnetron sputtering. The effect of sputtering DC power on the electrical, structural properties, and the nucleation and growth of Cu films during the initial stage of sputtering was investigated. The Cu films deposited at high sputtering power of 2.74 W cm$^{-2}$ showed strong crystallinity, low electrical resistivity of $1.86 \times 10^{-2}$ $\mu\Omega\cdot$m compared to the Cu films deposited at low sputtering power of 0.55 W cm$^{-2}$. From the results of AFM images, it was found that high sputtering DC power progresses the nucleation and the 3D island growth of Cu films without any mechanism change.

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REFERENCES