Influence of Substrate Bias Voltage on the Properties of Cu Thin Films by Sputter Type Ion Beam Deposition

Jae-Won Lim$^{1,*}$, Yukio Ishikawa$^{2}$, Kiyoshi Miyake$^{3}$, Mutsuo Yamashita$^{4}$ and Minoru Isshiki$^{1}$

$^{1}$Institute of Multidisciplinary Research for Advanced Materials, Tohoku University, Sendai 980-8577, Japan
$^{2}$Department of Substrate Engineering Semiconductor Division, Sumitomo Electric Industries Ltd., Itami 664-0016, Japan
$^{3}$Graduate School of Science and Engineering, Saitama University, Saitama 338-8570, Japan
$^{4}$Seinan Industries Co. Ltd., Osaka 559-0011, Japan

Cu thin films have been deposited on Si (100) substrate by using a non-mass-separated ion beam deposition (IBD) system. The effect of the substrate bias voltage on the properties of the deposited films was investigated using X-ray diffraction, resistivity measurement and field emission scanning electron microscopy. In the case of Cu thin films deposited without bias voltage, a columnar structure and small grains were observed distinctly, and the electrical resistivity of the deposited Cu films was very high. By increasing the bias voltage, no clear columnar structure and grain boundary were observed. The resistivity of Cu films decreased remarkably and at a bias voltage of $-50 \text{ V}$, reaching a minimum value of $18 \pm 1 \text{ n}\Omega \text{m}$, which is close to that of the bulk phase ($16.7 \text{ n}\Omega \text{m}$).

(Received January 24, 2002; Accepted April 30, 2002)

Keywords: ion beam deposition, copper, resistivity

1. Introduction

In ultra-large-scale integration (ULSI) structures, copper has been expected as a candidate for an interconnection material better than aluminum and aluminum-based alloys because of its low resistivity and excellent electromigration (EM) resistance. 1, 2) The use of Cu interconnections will improve device performance by decreasing the resistance-capacitance time delay and the power dissipation. However, the deposition of Cu thin films at relatively high temperature could result in undesirable diffusion of Cu into Si substrate and redistribution of impurities. 3) Besides, there still remain several problems which have to be solved for practical use of Cu films in deep submicron dimensions, such as (i) resistivity of Cu thin films higher than that of the bulk value, (ii) fast diffusion of Cu into Si substrate, (iii) reaction of Cu with Si at a relatively low temperature. 4) Among these problems, it is required to improve the resistivity of Cu thin films near to a bulk value, the others can be solved by inserting barrier layer between Cu film and Si substrate. Furthermore, to prevent the surface EM, that takes place via surface diffusion, it is particularly important to obtain clean and smooth surface. 5, 6) Therefore, there is a need for deposition technology preparing a high purity and low defect density Cu thin film for ULSI applications.

Mass-separated ion beam deposition (IBD) method is a useful technique to form high purity and low defect density films using ions of the deposit material and inert gas without a neutral atom component. 5-7) Although the mass-separated IBD method is important for basic research, it is not suitable for industrial applications because of small deposition area and low throughput. Non-mass-separated IBD method 8, 9) has been developed for industrial applications such as magnetic film devices, thin film transistor and Cu metal interconnec-

cations. In addition, a high purity RF sputter type ion source has been developed recently 5) for non-mass-separated ion beam deposition system. This ion source is composed of an RF (13.56 MHz) Cu coil and a high purity rod shape Cu target (99.9999%) located at the center of the RF coil. In our previous work, 8) Cu films, showing non-columnar structure and very smooth surface, were obtained by IBD system with this type of ion source. This smoothing and densification effect was attributed to irradiation of accelerated Cu$^+$ ions. Based on these results, the influence of the substrate bias voltage was examined in details in the present study. In addition, the purpose of this investigation is to optimize the proper negative bias voltage at which the Cu thin film has the smallest resistivity with good surface morphology.

2. Experimental

A schematic of a non-mass-separated IBD system with the RF sputter type ion source is shown in Fig. 1. The sputter type RF ion source consists of a Cu coil and a Cu target. The inner diameter of the RF copper coil (5 turns) is 57.5 mm. The target holder and the Cu coil were also water-cooled, and the Cu coil was connected to an impedance matching box for an RF (13.56 MHz) power supply. The chamber was evacuated by a turbo molecular pump and was maintained at a base pressure of $3 \times 10^{-4} \text{ Pa}$. A high purity (99.9999 mass%) Cu target is a rod shape of 8 mm in diameter and 150 mm in length. The Cu target was electrolytically polished to eliminate the surface contamination before loading and presputtered for 30 minutes to remove the impurities of the target surface prior to starting the deposition. The Si (100) substrates with the dimension of 18 mm $\times$ 18 mm were ultrasonically cleaned in ethanol, etched by 5% HF solution, and then loaded on the substrate holder through a load lock system. The substrate holders with the diameter of 50 mm were made of commercial 99.99% copper.
A high purity (99.9995%) argon gas was introduced into the chamber through a variable leak valve to maintain a fixed pressure. An Ar discharge plasma was generated at the fixed RF power with a reflected power of less than 40 W. When a negative DC bias voltage of −300 V was applied to the Cu target, the discharge plasma showed strong green emission as shown in Fig. 2, which suggested a creation of sputtered Cu atoms in excited states. When a deposition was going to begin, Cu$^+$ and Ar$^+$ ions were accelerated with a bias voltage between the discharge plasma and a Cu grid (20 mesh). The distance between the target and the substrate was changed from 35 to 60 mm and the negative DC bias voltage was in the range of 0 to −200 V. The deposition time was 900 s. The deposited area on the Si substrate was a dimension of 15 mm diameter. For the structural analysis of the films a Rigaku RINT 2000 X-ray diffractometer was used with Cu Kα radiation generated at 36 KV and 20 mA. Morphologies of the samples were observed with FE-SEM (Hitachi S-4100L). The deposition rates were obtained by measuring the film thickness by observation of SEM cross-section micrographs and the deposition time. Resistivity measurements of the samples were done by Van der Pauw method with indium electrodes.

3. Results and Discussion

In order to characterize the IBD system, relationship between the DC target current ($I_T$) and the DC target voltage ($V_T$) was measured at the first stage. Figure 3(a) and (b) show the DC target current ($I_T$) as a function of the DC target voltage ($V_T$) for various RF powers ($P_{RF}$) and Ar gas pressures ($P_{Ar}$), respectively. In both cases, the target current sharply increased up to about −20 V, and saturated at a target voltage of above −100 V. This characteristic is similar to that of mass separated IBD system reported by Yamashita. He demonstrated that the saturated target current density is more than one order of magnitude larger than that of the tetrode-type sputtering system. This saturated value of the target current is considered to be almost proportional to the plasma density, and it increased with increasing of $P_{RF}$ and $P_{Ar}$, as shown in...
Figs. 3(a) and (b). This fact suggests that the generation of the plasma required for the higher deposition rate is mainly caused and controlled by the RF energy applied to the RF coil and target voltage. However, in the present system, to prevent the melting of the Cu target at high RF power (over 240 W) and the target voltage (over −300 V), we selected experimental conditions that the RF power and the target voltage to be 240 W and −300 V, respectively.

Effect of Ar gas pressures on the deposition rate of Cu films was also examined. Figure 4 shows the deposition rate of Cu films and the Ar mean free path (MFP) change as a function of the Ar gas pressure. In general, the mean free path (λ) can be described by the following equation:11)

\[
\lambda = \frac{1}{\left(2^{1/2}\pi n \delta^2\right)}
\]

where \(n\) is a number of atoms per unit volume and \(\delta\) is a diameter of atom (Ar: 0.367 nm), \(n\) is given by eq. (2).

\[
n = \frac{P}{kT}
\]

Here \(P\) is gas pressure (Pa), \(k\) is an Avogadro’s constant and \(T\) is temperature (K).

The Ar MFP, calculated at room temperature, decreased gradually with increasing the Ar gas pressure, and the deposition rate of Cu thin films also decreased. Since a similar behavior must be applicable to a Cu MFP, the Cu MFP also decreases as the Ar MFP decreases. Thus, the Cu MFP decrease makes the Cu neutral particles and Cu⁺ ions ejected from the target become difficult to deposit on the substrate. Judging from above-mentioned results, the deposition rate can be independently controlled by changing either the RF power or the Ar pressure. Although the deposition rate increases with decreasing the Ar gas pressure, it is difficult in the present system to maintain the stability of discharge plasma operation for a long time at low Ar gas pressure of 9 Pa, we fixed the Ar gas pressure at 9 Pa.

In addition, in order to investigate the deposition rate of Cu films at various distance, we changed the target-substrate distance from 35 to 60 mm. In the present system, we could not decrease the distance less than 35 mm because of an existence of a shutter located in the deposition chamber. Figures 5(a) and (b) show the effect of the distance on the deposition rate and the resistivity, respectively. The deposition rate was found to increase noticeably with decreasing the distance. In particular, the deposition rate showed the highest value when the distance was 35 nm. The resistivity of Cu films gradually decreased with decreasing the distance as shown in Fig. 5(b). This value was much higher than that of the bulk value. Therefore, the substrate bias voltage was applied at 35 mm of the target-substrate distance for a better improvement of the electrical property of Cu films.

The deposition rate of Cu films was inversely proportional to the negative substrate bias voltage as shown in Fig. 6(a). The decrease of the deposition rate must be related to a film density and sputtering caused by ion bombardment on the surface. Recently, Choi et al.12) reported that as the substrate bias voltage increased, the Cu film density increased and saturated to nearly bulk value at a voltage of −100 V. This can explain the decrease of the resistivity with increasing the bias voltage in our experimental results. It is found that the resistivity of Cu films decreased drastically and reached the minimum value of 18 ± 1 nΩm at a bias voltage of −50 V, which was very close to bulk value of 16.7 nΩm. In our recent study, the resistivity of Cu films deposited at the substrate bias voltage of −50 V have maintained values of around 18 nΩm in the case of thickness over 100 nm. Therefore, the resistivity of films was independent on the thickness in present results because the deposited samples have the thickness over 100 nm. Further increase of the bias voltage up to −200 V, results in gradually increasing the resistivity as shown in Fig. 6(b). The increase of the resistivity at higher bias voltages

![Fig. 4 Deposition rate of Cu films as a function of the Ar gas pressure (P_{RF}: 240 W, V_T: −300 V, D_{TS}: 50 mm).](image)

![Fig. 5 Deposition rate (a) and the resistivity (b) as a function of the target-substrate distance (P_{RF}: 240 W, V_T: −300 V, P_{Ar}: 9 Pa).](image)
could be explained by the excessive resputtering of ion bombardment on the surface. Although a similar resistivity behavior was also reported for bias sputtered Cu films by other investigators,\textsuperscript{12,13} the minimum resistivity of Cu films, reported so far, was larger than 20 n\(\Omega\)m, which is still higher than 16.7 n\(\Omega\)m of the bulk value. It is worth noting that the resistivity of Cu films deposited by IBD system with the high purity ion source has a nearly bulk value at the bias voltage of −50 V.

To examine the effect of the bias voltage on the surface morphology and the texture of Cu films, we carried out SEM observation of the samples at bias voltages of 0, −50, −100, −150 and −200 V. Figures 7 and 8 show the effect of the negative DC bias voltage on the surface and the cross-sectional morphologies of the same samples, respectively. As shown in Figs. 7(a) and 8(a), in the case of the films deposited without bias voltage, which means that the deposition is mainly caused to Cu neutral particles and the surface diffusion is insufficient, Cu film had the columnar structure with small grains as often observed in sputter deposition. On the other hand, when the negative bias voltage of −50 V was applied to the substrate, the columnar structure was not observed any more, the deposition is thought to be due to both of Cu neutral particles and accelerated Cu\(^+\) ions. This indicates that the effect of Cu\(^+\) ions bombardment with sufficient energy enhances the surface migration during the deposition.

The presence of the ionized particles, even when only a few percents of ionized particles are included, to be considered to influence greatly the critical film property of deposited mate-
Hirsch et al. investigated the effect of ion bombardment and came to conclusion that when part of the surface was bombarded by argon ions on the germanium films, the deposit showed stronger bonding between the deposited film and the substrate, and no sign of flaking anywhere on the irradiated area was observed. Evidence of the effect of ion bombardment also can be seen in the case of SEM morphology as shown in Figs. 7 and 8. That is the reason why clear grains, grain boundary and columnar structure were not observed and the roughness of the surface decreased remarkably as shown in Figs. 7(b) and 8(b).

However, applying the bias voltage of $-100$ V increases the grain size, and the surface of the Cu film become slightly rougher as shown in Figs. 7(c), 8(c). The resputtering of the surface caused by the higher energy particles makes the surface rougher during the deposition. Further increasing the bias voltage, although the grain size was not changed, the surface of the Cu film became much rougher as shown in Figs. 7(d), (e), 8(d), (e). Morphology change with the bias voltage is in good agreement with the result expected from the resistivity measurement as discussed above. Thus, it is concluded that the optimum negative bias voltage is $-50$ V, which results in the smallest resistivity with good surface morphology.

Figure 9 shows X-ray diffraction patterns recorded by $\theta$–$2\theta$ scanning in a range of $40^\circ$ to $55^\circ$. The dominant Cu (111) orientation is observed regardless of the substrate bias voltage. The intensity of the Cu (111) peak was strengthened extremely when the bias voltage of $-50$ V was applied to the substrate, although the appearance of the Cu (200) peak was observed. From the XRD patterns, a ratio of relative peak intensities $I_{(111)}/I_{(200)}$ was estimated and shown in Fig. 10. Considering that, the ratio for randomly oriented Cu powder is $I_{(111)}/I_{(200)} = 2.17$, a strong (111) texture was obtained in these samples when the bias voltage was applied. The ratio of $I_{(111)}/I_{(200)}$ increased with increasing negative bias voltage until it reached the bias voltage of $-150$ V, and then decreased at a bias voltage of $-200$ V as shown in Fig. 10. Although the ratio of $I_{(111)}/I_{(200)}$ was high in the case of the bias voltages of $-100$ and $-150$ V, it seemed that it is undesirable to be inferior to other properties compared with the
Median Time to Failure (MTF) is proportional to the strong stress as shown in Fig. 11. Choi et al. showed that the residual stress is related to the substrate bias voltage. The Cu film deposited without bias voltage is in a state of high tensile stress. With increasing bias voltage, the tensile stress decreases and saturates to nearly zero at a bias voltage of −50 V. Therefore, the decrease of the FWHM value of Cu (111) reflection peak at the bias voltage of −50 V is due to the decrease of the residual stress, caused by sufficient energy which enhances the surface migration. However, the increase of the FWHM value of Cu (111) peaks, in the case of further increase the bias voltage up to −200 V, seems due to formed defects caused by excessive higher energy particle bombardment on the surface during deposition. This behavior of FWHM showed excellent agreement with changes of the resistivity and morphology observation.

4. Conclusion

Cu thin films have been deposited by using the non-mass-separated RF sputter type IBD system on Si (100) substrate at various bias voltages. An optimum applied negative bias voltage of −50 V was found to prepare Cu films with no columnar structure, the lowest electrical resistivity (18 ± 1 nΩm) and good surface morphology. The lowest FWHM value of the Cu (111) reflection peak at the bias voltage of −50 V corresponded to the minimum resistivity value as well as smooth surface and good morphology. These results indicate that the IBD system developed in the present study is suitable to fabricate Cu thin films for ULSI applications.

REFERENCES