Estimations of Plasma Parameters and Bias-Sputtering of Cu Films in Gas-Flow-Sputtering Operating at 1-Torr Pressure Region

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Gas flow sputtering operates at a high pressure ranging from 0.2 Torr to 10 Torr, which enables a "soft" film growth process and good step coverage. In GFS, bias-sputtering, which induces ion bombardment of growing films, was found to be very useful to obtain Cu films with a low resistivity. The optimum bias voltage was -60 V and low resistivity of 2.6 $\mu\Omega$ cm, which is close to the bulk value (1.72 $\mu\Omega$ cm), was achieved. The bias-sputtering also resulted in the grain growth of fcc crystallite of Cu. A probe measurement revealed that stable and high density plasma of the order of 10¹³ cm⁻³, which is two orders higher than that of conventional sputtering method, was produced in the hollow target (a Cu tube of 4 cm in diameter and 6 cm in length) and was transported by a directed Ar flow to the substrate. The stable and effective bias-sputtering in GFS is attributed to this high density of plasma with potential of a few eV against the ground. These results show that bias-sputtering in GFS is useful at a low voltage around -60 V because of high operating pressure and high density of plasma.

Key words: sputtering, gas flow sputtering, bias sputtering, sputtered film, Cu film

1. INTRODUCTION

Sputtering is one of useful physical vapor deposition (PVD) methods in various film applications and is used for the fabrication of interconnects such as Al allov or Cu films with low resistivities in semiconductor devices or microwave devices [1]. However, it may give rise to serious trouble in the reliability of the next generation of highly integrated circuits because of its poor step coverage. We have developed a sputtering method, termed gas flow sputtering (GFS) [2], which operates at a high pressure above 0.2 Torr, resulting in the improvement of step coverage [3], and we have recently found that Cu films with a low resistivity near the bulk value can be obtained by GFS with an addition of ion bombardment during the film growth [4]. In GFS process, since a high density of discharge plasma penetrates near the substrate, an ion bombardment to the growing film is efficiently added by the technique of bias-sputtering, i.e., the addition of negative voltage to the substrate during the sputtering.

GFS is based on a hollow cathode discharge and is schematically shown in Fig. 1. A tube or a couple of facing plates is used as the target, in which hollow cathode discharge occurs. The most characteristic feature of GFS is a high operating pressure ranging from 0.2 Torr to 10 Torr, which results in the complete thermalization of energetic particles including sputtered particles to the temperature of sputtering gas (Ar) by collisions with Ar [5]. The thermalized particles are transported by the directed flow of Ar and deposited on the substrate. As the result, GFS enables a "soft" film growth process and good step coverage similar to that of chemical vapor deposition (CVD).

One of other advantages of GFS is that a reactive sputtering of metal mode with a high deposition rate is possible by supplying the reactive gas (e.g., oxygen) in front of the substrate, because the reactive gas cannot penetrate the target due to Ar gas counter flow [6]. In GFS, the discharge plasma is also transported by Ar flow to the deposition region, but its behavior has never been investigated.

In this study, plasma parameters of discharge plasma near the substrate were evaluated by a probe method in order to obtain more knowledge about the biassputtering in GFS. In addition, the effects of biassputtering on the resistivity of Cu films were investigated.



Fig.1 Principle of gas flow sputtering.

2. EXPERIMENTAL PROCEDURE

2.1 Estimation of plasma parameters

The plasma parameters such as the electron density, the electron temperature, and the space potential, were estimated from the voltage-current characteristics measured by a standard Langmuir probe method. The probe had a cylindrical shape and the electrode was a tungsten wire with the diameter of 0.1, 0.3 or 0.6 mm and length of 0.5, 1.0 or 7.0 mm. The experimental set-up is shown in Fig. 2; in this paper, the measuring points are presented by the coordinate shown in this figure. The target was a Cu tube with purity of 99.99%, and its internal diameter and length was 4 cm and 6 cm, respectively. The sputtering gas was 99.9999% Ar and its flow rate was 500 sccm. The apparatus and its operation are described elsewhere [7].



Fig. 2 A schematic drawing of the experimental set-up and the coordinate system to measure a probe characteristics of plasma.

2.2 Bias-sputtering of Cu films

Deposition of Cu films was performed with Ar flow rate of 500 sccm, total pressure of 1.0 Torr and dc discharge power of 580 W. The deposition rate for target-to-substrate separation (S-T) of 5 cm and 8 cm was 45 nm/min and 9 nm/min, respectively. Glass plates were used as the substrate. Al films were coated at the edges of substrate, and negative dc bias voltage was applied through the Al films. Therefore, dc bias was effective after the Cu films became electrically continuous, maybe over 10 nm in thickness. The thickness of specimen films was 200 nm and, thus the initial layer without ion bombardment is neglected in the evaluation of effects for bias-sputtering. The electrical resistivity of specimen was measured at room temperature by a standard four probe method. The crystal structure was characterized by x-ray diffraction (XRD).

3. RESULTS AND DISCUSSION 3.1 Discharge characteristics

The plasma was generated by a dc discharge between the anode and the cathode (target). A current-voltage characteristic curve of the discharge is shown in Fig. 3. The so-called hollow cathode effect is seen to be effective above 0.4 Torr, that is, the discharge is maintained by a relatively low voltage and large current is obtained. It is a very desirable feature that the voltage maintaining discharge is not so low but around -300 V, because the cathode voltage ranging from -300 V to -600 V is suitable for sputtering [8].





3.2 Plasma parameters

Typical plasma parameters under a condition of 1.0 Torr of Ar gas with different flow rates are shown in Fig. 4. The flow rate of the gas is set to be 0 (closed circles in Fig. 4), 500 (open circles), and 1000 (mark x's) sccm. These parameters were not affected by a substrate voltage.

First of all, it is clear from the figure that a quiet (i.e., low electron temperature) and stable plasma, which is indispensable for film growth, is generated. The electron temperature, T_e in Fig. 4 (a), is approximately 0.2 - 0.3 eV. Its spatial profile along the *x*-axis is almost constant and uniform even when the gas flow rate is different.

The electron density, n_e in Fig. 4 (b), is order of 10^{13} cm⁻³ around x = 0 (cm). The value of the density is two orders higher than the conventional sputtering method. Such a high current discharge results in very high sputter-deposition rates. The electron density decreases with increasing distance, x. The electron density profile is approximated as follows:

$$\log n_e = \begin{cases} -0.50 \ x + 0.35 \ (0 \ \text{sccm}), \\ -0.26 \ x + 0.99 \ (500 \ \text{sccm}), \\ -0.15 \ x + 0.80 \ (1000 \ \text{sccm}), \end{cases}$$

where x and n_e are measured in units of cm and 10^{13} cm⁻³, respectively. It is found that a gradient of the density reduction depends on the gas flow. In the case of 0 sccm, there is no gas flow and the plasma only diffuses from its generated region. The gradient becomes more flat when the gas flow speed is higher. This suggests that the plasma is carried by the gas flow with diffusing toward perpendicular direction to the x-axis.



Fig. 4 The electron temperature profile (a), the electron density profile (b), and the space potential profile (c).

The space potential, φ_s , for 0, 500, and 1000 sccm are approximately 1, 1.8, and 3 eV in Fig. 4 (c). Except for the case of 0 sccm, where the space potential gradually decreases, the space potential is almost constant. **3.3 Resistivity of Cu films** Figure 5 shows the resistivities of Cu films obtained by bias-sputtering with various bias voltages. The resistivity ρ decreases with increasing bias voltage to -60 V and increases above the voltage. This tendency is seen for both cases of S-T=5 cm and S-T=8 cm; however, the resistivity for the case of S-T=5 cm is much lower than that of S-T=8 cm. For the sample obtained at S-T=5 cm and bias voltage of -60 V, minimum resistivity with value of 2.6 $\mu\Omega$ cm is achieved, which is close to the bulk resistivity of 1.72 $\mu\Omega$ cm.

Figure 6 shows XRD patterns of Cu films obtained with various bias voltages. The diffraction peaks become strong in intensity with increasing the bias voltage, but they become weak at -80 V and become strong again above that voltage. The out-of-plane coherence length of crystallites (crystallite size normal to the film plane) was evaluated from the peak width of diffraction peak using Scherer's equation [9] for every sample. The results are shown in Fig. 7. The comparison of the bias dependencies between the coherence length shown in this figure and ρ shown in Fig. 5 suggests that the ion bombardment by bias-sputtering promotes the growth of crystallites and, as the result, the reduction of film resistivity takes place. However, since the resistivity of metals is dominated by electron scattering factors such as phonons, grain boundaries, impurities, structural defects etc., more detailed experiments are necessary to clarify the main origin of resistivity reduction by bias-sputtering.



Fig. 5 Dependence of resistivity on the bias voltage for bias-sputtered Cu films in GFS.

Comparing the cases between S-T=5 cm and S-T=8 cm, lower resistive films are deposited at 5 cm rather than at 8 cm and the electron density at 5 cm is approximately four times larger than that at 8 cm for 500 sccm, which reveal that higher density of plasma is more beneficial to bias-sputtering. On the other hand, minimum resistivity is obtained at bias voltage of -60V

in both cases. Although the effects of ion bombardment must depend on the both energy and number of bombardment ions, this tendency suggests the ion energy is more important to promote crystal growth and to reduce the resistivity.

The voltage dependencies of resistivity and coherence length of crystallites at the region of bias voltages above 60 V, shown in Figs. 5 and 7, are interesting. However, the origins of the behavior are not well understood now.

4. SUMMARY

It was found that a stable and high density plasma of the order of 10^{13} cm⁻³ was produced in the hollow Cu tube and was transported by Ar flow to the deposition region. The relatively low voltage around -300 V was needed for maintaining the discharge, which is suitable for a sputtering phenomena. The electron density decreased with increasing distance from the cathode due to the diffusion toward the perpendicular direction. In the deposition of Cu films by GFS, bias-sputtering with about -60 V showed marked effects for the reduction of resistivity with minimum value of 2.6 $\mu\Omega$ cm and also for the crystal growth.

It is considered that this gas-flow-sputter method is suitable not only for the deposition of low resistive Cu films but for a dry-cupper-plating to a small-sized structure such as a thin wire mesh, because sputtered particles and a high density of plasma are transported by a forced atmospheric current of Ar.



Fig. 6 XRD patterns of Cu films obtained by bias-sputtering with various bias voltages.



Fig.7 Dependence of out-of-plane coherence length of crystallites on the bias voltage for bias-sputtered Cu films in GFS.

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(Received December 23, 2004; Accepted January 31, 2005)