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Effect of Negative Substrate Bias Voltage on the Nucleation and Growth of Cu Films during the Initial Stage of Ion Beam Deposition

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We focused 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 resultant microstructure of Cu films was observed by atomic force microscopy (AFM). It was found that Cu films with or without a negative substrate bias voltage have a different dependence of nucleation and growth. It was established that the mechanism of nucleation and growth of Cu films is changed to a progressive nucleation and lateral growth by a sufficient migration of adatoms accelerated by applying a negative substrate bias voltage.

Keywords: copper, nucleation, growth, substrate bias voltage, atomic force microscopy

1. INTRODUCTION

In ultra large-scale integrated circuits, the continuing demand for a lower resistivity of metal interconnects has led to the choice of copper as an alternative to aluminum because copper offers a bulk resistivity (1.67 mΩcm) that is lower than aluminum resistivity (2.67 mΩcm) [1,2]. It is widely known that the application of a negative bias voltage to the substrate can enhance characteristics (magnetization, resistivity, microstructure, crystallographic orientation etc.) of thin films prepared by a sputtering or deposition process [3-8]. We have also reported the effect of the negative bias voltage on various properties of Cu films deposited using ion beam deposition [9-12]. However, most studies have paid no attention to the effect of the negative substrate bias voltage on the nucleation and growth of Cu films during the initial stage of deposition. Therefore, the nucleation and film growth of Cu films with or without applying a negative substrate bias voltage and their relations to the resultant microstructure were evaluated by atomic force microscopy (AFM).

2. EXPERIMENTAL PROCEDURE

Cu films were deposited by non-mass separated ion beam deposition (IBD), the schematic diagram of which has been reported elsewhere [9]. The IBD apparatus consists of a deposition chamber and a load-lock system. The ion source, which is composed of an RF (13.56 MHz) Cu coil and a Cu target, is located in the deposition chamber (base pressure of 10^{-5} Pa). The Cu target was electrochemically polished to eliminate surface contamination, then pre-sputtered for 30 minutes to remove the surface contaminated layer prior to starting the deposition. Si (100) substrates were ultrasonically cleaned in acetone and etched by 5 % HF solution. The substrate holders with a diameter of 50 mm were made from commercial 99.99 % copper. High purity (99.9995 %) argon gas was injected into the deposition chamber to form argon plasma generated by 260 W with a reflected power of less than 40 W. Cu plasma was generated by an application of a negative bias voltage of –300 V to the Cu target. Cu films were deposited at a substrate bias voltage of 0 V and –50 V (hereafter Cu films Vs = 0 V and –50 V) under Ar pressure of 9 Pa for a different deposition time. It was demonstrated in our previous work [9] that Cu films at Vs = –50 V have the lowest resistivity and a good surface morphology. The distance between the target and the substrate was 35 mm. The microstructure and the morphology of the Cu films were observed using a field emission secondary emission microscopy (FE-SEM: HITACHI S-4100L). AFM (SII SPA 300) analysis was performed to evaluate nucleation and growth of Cu films.

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3. RESULTS AND DISCUSSION

Figure 1 shows surface and cross-sectional SEM observations for the Cu films at $Vs = 0$ V and –50 V. As seen in Figs. 1(a) and 1(c), the Cu film at $Vs = 0$ V have surface grooves between the small grains. A columnar structure is distinctly seen from the interface to the surface in the cross-sectional observation. On the contrary, no visible columnar structure is observed for the Cu film at $Vs = -50$ V. Furthermore, it was found that the average grain size of the Cu film at $Vs = -50$ V is much larger than that of the Cu film at $Vs = 0$ V. We reported in the previous study [13] that the average grain size of the Cu film at $Vs = -50$ V is 2.3 times larger than that of the Cu films at $Vs = 0$ V. Yamamoto and Ichimura [14] presented morphology changes of TiN films according to the negative substrate bias voltage. They reported that at a low substrate bias voltage, the columnar units grown outward from the interface, while the film morphology changed to a much denser microstructure at higher substrate bias voltage above –200 V. The result was explained by an effective enhancement in adatom diffusivity and enhanced nucleation kinetics. Hirsch and Varga [15] also proved the similar effect by the ion bombardment on germanium films. These serve to explain the reason why the grains and the columnar structure were not appeared, and the roughness of the surface decreased markedly on the films deposited at the substrate bias voltage.

In the present study, to investigate a difference of the nucleation and growth between two types of Cu films during the initial stage of ion beam deposition and resultant microstructure, we tried to observe directly the nucleation and growth of Cu films at $Vs = 0$ V and –50 V as a function of deposition time. Figures 2 and 3 show 3D and 2D AFM images of the Cu films at $Vs = 0$ V and –50 V for ~1 s, 5 s, 10 s, 30 s, and 60 s, respectively. The thicknesses of the Cu films at $Vs = 0$ V and –50 V for 60 s are approximately 28 nm and 24 nm, respectively. The AFM images reveal that there is a different nucleation and growth mechanism between two types of the Cu films. In the initial nucleation stage, there is a distinct difference between two types of the Cu films. The main difference is that the number of nuclei on the substrate

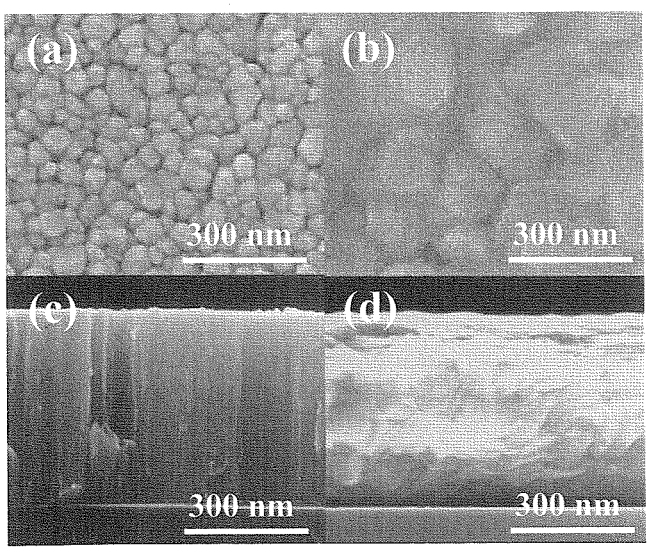

Fig. 1. Surface and cross-sectional SEM micrographs of the Cu films: (a) and (c) $Vs = 0$ V, (b) and (d) $Vs = -50$ V.

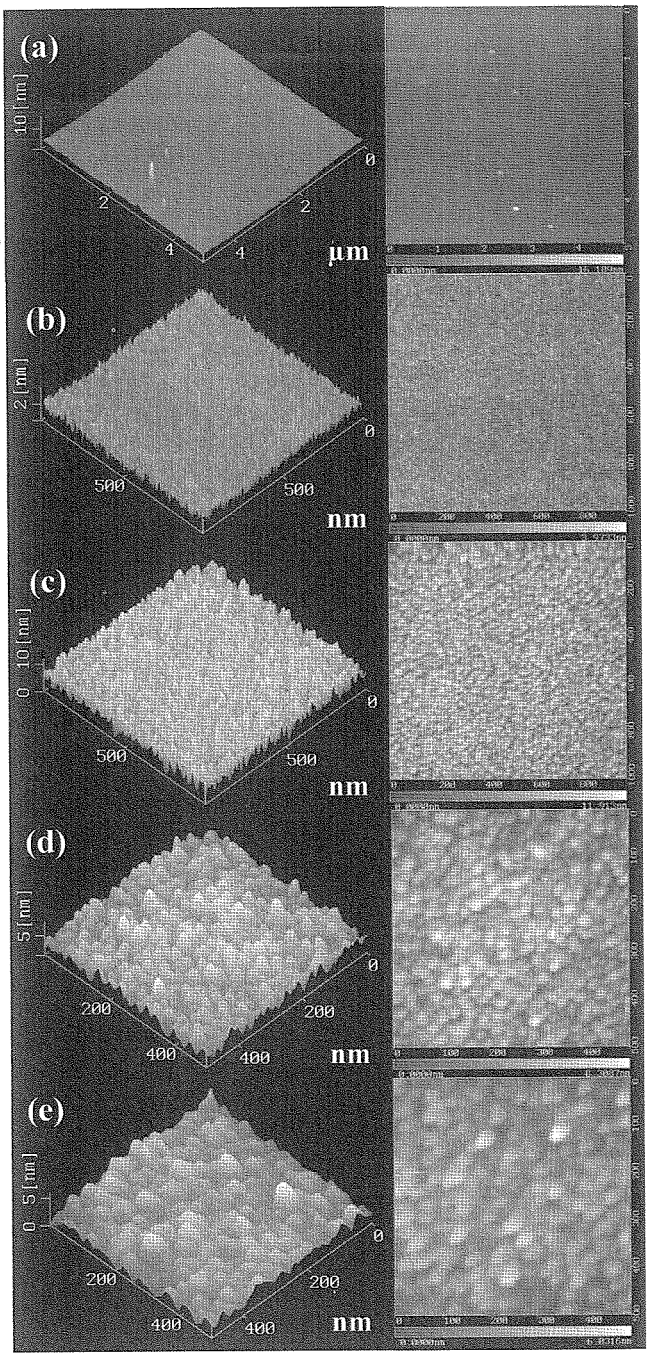

Fig. 2. 3D and 2D AFM images of the Cu films at $Vs = 0$ V as a function of deposition time: (a) ~1 s, (b) 5 s, (c) 10 s, (d) 30 s, and (e) 60 s.

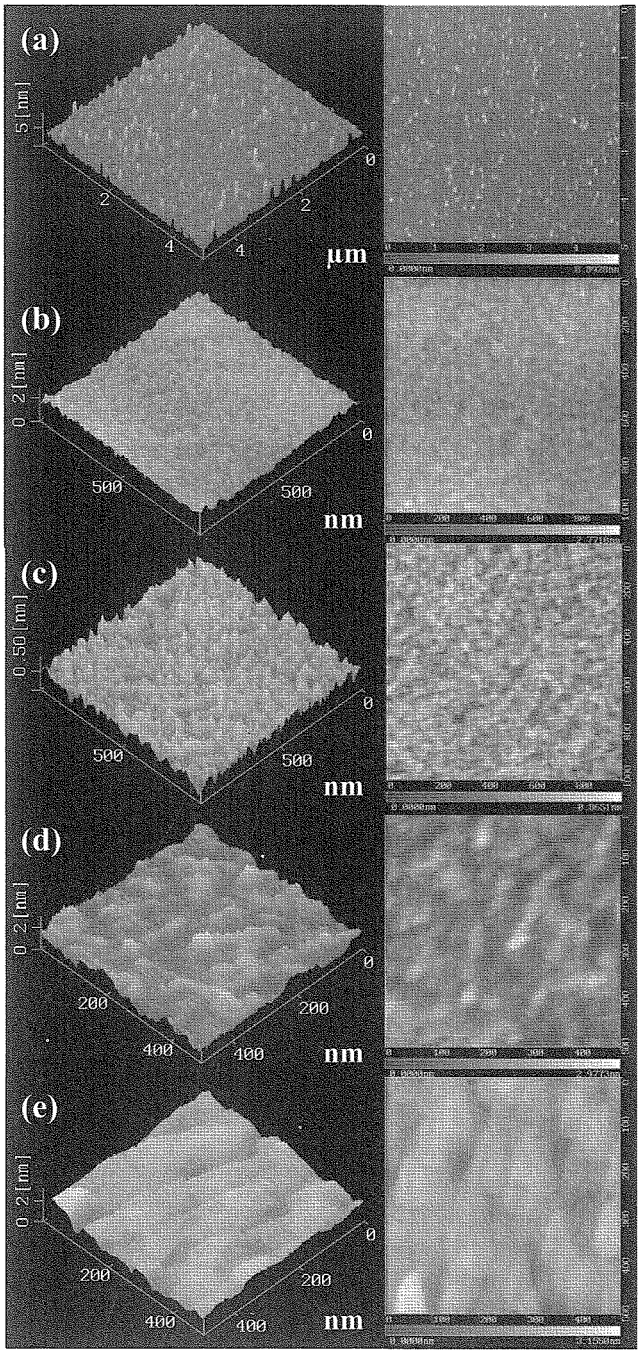

Fig. 3. 3D and 2D AFM images of the Cu films at Vs = –50 V as a function of deposition time: (a) ~1 s, (b) 5 s, (c) 10 s, (d) 30 s, and (e) 60 s.

for the Cu film at Vs = –50 V after ~ 1 s is much greater than those on the substrate for the Cu film at Vs = 0 V. That is, only several nuclei on the substrate are occurred after ~1 s deposition time for the Cu films at Vs = 0 V (Fig. 2(a)), whereas, the number of nuclei are already formed after ~1 s for the Cu film at Vs = –50 V. Besides, it appears a different behavior on the grain formation and growth between them. For the Cu film at Vs = 0 V, Cu grains are formed and grown through coalescence of the nuclei as seen in Figs. 2(b) to (e), which represents a typical island growth and finally the columnar structure, as shown in Fig. 1(a), are formed. On the other hand, as shown in Fig. 3(a), due to the enhanced migration of adatom on the substrate by a negative substrate bias voltage, the nuclei observed initially tend to change into a lateral growth during grain formation. Therefore, the surface of grains grown broadly becomes very smooth through grain coalescence, which results in large grains and a flat surface (see Figs. 3(b) to (e)).

RMS roughness values of the Cu films at Vs = 0 V and –50 V from a 1 mm × 1 mm surface area were also investigated and are plotted in Fig. 4. The Si substrate had a very flat surface as a RMS value of 0.07 nm. We excluded the value of the Cu films at the deposition time of 1 s due to very small nucleus formation. For the Cu films at Vs = 0 V, the RMS value increases significantly in the deposition time from 5 s to 10 s, and decreases slightly to approximately 1.0 nm. On the other hand, the RMS value of the Cu films at Vs = –50 V initially increases slightly at 10 s and decreases and retains a low value of about 0.3 nm.

There are three basic modes of growth based on the principal interactions between substrate atoms and deposited atoms [16]; (a) the layer growth (2D), (b) the island growth (3D), and (c) the Stranski-Krastanov growth (3D islands on the top of one or a few layers. In our case, the growth mode of the Cu films at Vs = 0 V corresponds to the 3D island growth, whereas the growth mode of the Cu films at Vs = –50 V seems to be changed from the 3D island growth to the lateral growth from the deposition time of 10 s. It is well known that the kinetic energy of atoms toward the substrate has an influence on the nucleation rate and nuclei density increase [17,18], which can be generated in this study by the bombardment with high energy particles accelerated by applying a negative substrate bias voltage. Therefore, the mechanism

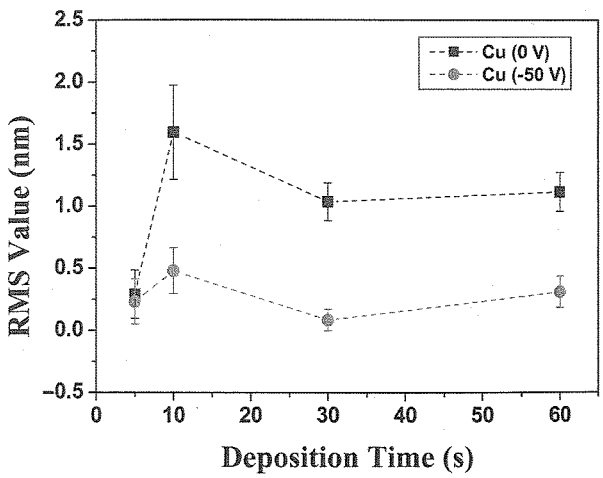

Fig. 4. Surface RMS values of the Cu films at Vs = 0 V and –50 V as a function of deposition time.

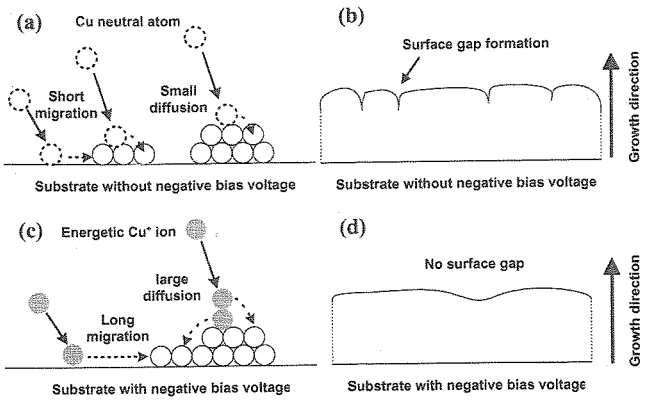

Fig. 5. Schematic illustrations of nucleation and film growth: (a) and (b) without a substrate bias voltage, (c) and (d) with a substrate bias voltage.

of the nucleation and growth seems to be changed to progressive nucleation and lateral growth by sufficient migration of adatoms accelerated by a negative substrate bias voltage, which results also in an improvement of film morphology. From the results of the AFM images and the RMS values, Cu films with or without a negative substrate bias voltage are found to have a different dependence of nucleation and growth.

Concerning the mechanism of the nucleation and film growth on the substrate, the movement of adatoms and resulting morphology of Cu films on the substrate with or without a negative substrate bias voltage are described in Fig. 5. As shown in Figs. 5(a) and (b), for the substrate without a negative bias voltage, the 3D island growth seems to be due to insufficient surface migration and diffusion of low energy particles, which are considered mainly to be composed of Cu neutral atoms. These insufficient surface migration and diffusion of Cu atoms cause a columnar structure with small grains. Besides, as the thin film grows, many gaps between small grains form on the surface. In contrast, when the substrate bias voltage is applied (see Figs. 5(c) and (d)), a large amount of Cu^+ ions with high kinetic energies bombard the substrate. Then incident momentum of the Cu^+ ions is transformed into sufficient surface migration and diffusion energies of each atom. For this reason, a lateral growth of grains can be occurred, which lead to a comparatively uniform and smooth morphology after the film growth.

4. CONCLUSION

The effect of a negative substrate bias voltage on the nucleation and growth of Cu films during the initial stage of ion beam deposition and their relation to the resultant microstructure were observed. Experimental results indicated that Cu films with or without a negative substrate bias voltage are found to have a different dependence of nucleation and growth. The mechanism of nucleation and growth of Cu films is changed to a progressive nucleation and lateral growth by a sufficient migration of adatoms accelerated by applying a negative substrate bias voltage.

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