Grain Size Distribution at the Bottom Region in Very Narrow Cu Interconnects
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ABSTRACT
Crystal grain sizes less than 40 nm diameter, i.e., mean free path of an electron in Cu, in both the upper and bottom part of Cu interconnects with 70 nm width and 150 nm depth as a function of overburden Cu plated films with 150 nm, 300 nm and 450 nm thickness have been evaluated by X-ray diffraction method. In the upper part of the Cu interconnects, the frequency ratio of the crystal grains with less than 40 nm diameter was quite small and changed little in the Cu interconnects among overburden Cu films with different thickness. In the bottom part of the Cu interconnect, however, the frequency ratio of the crystal grain with less than 40 nm diameter was found to be very dependent on the thickness of overburden Cu films. The frequency ratio was about 20% at the overburden Cu film thickness of 150 nm, while the frequency ratio decreased to less than 2% at the overburden Cu film thickness of 450 nm. The resistivity of narrow Cu interconnects was found to increase with the increase of the frequency ratio of crystal grains less than 40 nm diameter at the bottom part of Cu interconnects.

1. Introduction

Very narrow Cu interconnects are indispensable for high performance ULSIs. However, the resistivity of narrow Cu interconnects increases drastically with decreasing the interconnect line width and that leads to performance degradation of ULSIs. The resistivity increase in narrow Cu interconnects is mainly due to the fact that electron scattering at very small grain boundaries increases with decreasing grain sizes. In addition, widely dispersed small grains deteriorate electro-migration resistance in Cu interconnects. The uniform coarsening of the crystal grains in very narrow interconnects is considered to be one of the crucial points to be tackled for development of future high performance ULSIs.

As factors affecting the crystal grain size in electroplated Cu interconnects, the electroplating conditions, such as overburden Cu film thickness and purities of the electrolyte solution and anode, and the interconnect trench geometry, i.e., its width and depth, have been pointed out. During annealing of Cu interconnects, grain growth has been reported to originate in the overburden Cu film and to extend the Cu interconnect in the trench. It was reported that grains in narrow Cu interconnects did not grow well during annealing due to the strong restriction from both walls of the trench and this in narrow Cu interconnects did not grow well during annealing due to the strong restriction from both walls of the trench and this in narrow Cu interconnects did not grow well during annealing due to the strong restriction from both walls of the trench.

On the other hand, it has been reported that the grain size in the bottom part of the annealed Cu interconnect is significantly smaller than that at the upper part. However, previous reports dealt with the relationship between mean grain size throughout the depth direction of a interconnect and resistivity. Hence, mean grain size does not reflect the resistivity increase necessarily. It is very important to clarify the effect of smaller grain sizes at the bottom part of interconnects on the resistivity of Cu interconnects for manufacturing low resistance interconnects.

In this research, we used an X-ray diffraction method to evaluate the grain size distributions in the 70 nm wide and 150 nm deep Cu interconnects. The merit of the X-ray diffraction method is its capability to easily obtain the grain size distributions including the grains with less than 40 nm diameter, i.e., mean free path of an electron in Cu. We also evaluated the grain size distributions in the depth direction by measuring the Cu interconnect polished to nearly half the trench depth. We studied the effect of thickness of overburden films on the grain size distributions at the bottom part of Cu interconnects in order to estimate the impact of smaller crystal grains less than 40 nm at the bottom part on the resistivity of interconnects.

2. Experimental Procedure

Cu interconnects were made by DC electroplating. Trenches for 70 nm wide and 150 nm deep interconnects were made using electron beam lithography and reactive ion etching in a SiO2 layer on a 10 mm square SiO2/Ti/Si substrate. The trenches were made on a substrate such that the Cu interconnects in the substrate have sufficient X-ray diffraction peak intensity for analyzing the grain size distribution.

The Ultrathin Ta/N/Ta (TaN, 7.5 nm; Ta, 7.5 nm) barriers and a 50 nm thick Cu seed layer were sputter-deposited in that order. Schematic drawings of a trench structure and the spacing of each trench in the substrate for X-ray diffraction measurement are shown in Figs. 1(a) and (b), respectively. The DC electroplating was done at room temperature with a current density of 5 mA/cm² using a high purity 99.9999% (6N) Cu anode and 6N CuSO4·5H₂O electrolyte with additives. The electroplated overburden Cu films were 150, 300 and 450 nm thick. Cu interconnect specimens were annealed in the IR-RTA equipment at 573 K for 10 min in vacuum (5 × 10⁻⁵ Torr). After that, the overburden Cu film and TaN/Ta barriers were removed by chemical mechanical polishing (CMP).
Furthermore, several specimens were polished to get bottom half interconnects (about 80–100 nm height) by CMP. The heights of polished Cu interconnects were confirmed by cross-sectional SEM observation.

The X-ray diffraction patterns of the Cu interconnect specimens made on a 10 nm square SiO$_2$/Si substrate were measured with a diffractometer using CuK$_\alpha$ radiation. The angular step of 20 and the counting time were 0.02° and 1.0–80.0 s, respectively. After determining area- and volume-weighted average column lengths from diffraction peak profiles using the Fourier method of Warren-Averbach,12 grain size distribution in the Cu interconnect was estimated from two average column lengths by assuming spherical grains and their log-normal size distribution.13,14

3. Results and Discussion

Figure 2(a) shows a cross-sectional SEM image of the Cu interconnect specimen, in which the 450 nm thick overburden Cu film and TaN/Ta barriers were polished by CMP after annealing. We call this Cu interconnect hereinafter the whole interconnect. Figure 2(b) shows the Cu interconnect specimen polished to nearly half the trench depth in addition to the overburden Cu film and TaN/Ta barriers. We call the Cu interconnect thus polished as the bottom half interconnect. From the SEM images, the depth of the whole and bottom half interconnects are estimated to be 153 and 100 nm, respectively. The depth ratio of the former to the latter is calculated to be 0.65.

Figure 3(a) shows the X-ray diffraction pattern of the whole interconnect. Clearly the whole Cu interconnect has a strong (111) texture which is the same as reported previously for different width Cu interconnects.9,11 The accurate (222) peak profile shown in the inset was measured using a much longer counting time. As shown in Fig. 3(b), the strong (111) texture is still maintained in the bottom half interconnect. The (111) peak profile from the bottom half interconnect is measured at nearly half the intensity compared with that from the whole interconnect. We estimated the integrated intensity of (111) reflection on the whole and the bottom half interconnects. The integrated intensity ratio of the former to the latter is 0.63. Since the X-ray irradiation area in both interconnect specimens are nearly the same, this integrated intensity ratio is considered to correspond to the depth ratio in these interconnects. The depth ratio obtained from the integrated intensity of (111) reflection is almost the same as that obtained from the SEM images shown in Fig. 2. Therefore, the grain size distribution in the whole and bottom half interconnects were represented taking into account the depth ratio or the integrated intensity ratio.

The grain size distributions in nanocrystalline materials have been reported to be well described by a log-normal function.13 The log-normal distribution $g_{LN}(D)$ of the grain diameter $D$ has the form

$$g_{LN}(D) = \frac{1}{\sqrt{2\pi D \ln \sigma}} \exp\left(-\frac{(\ln(D/D_0))^2}{2(\ln \sigma)^2}\right)$$  \hspace{1cm} (1)

where $D_0$ and $\sigma$ are the median diameter and the relative width, which is a parameter related to the width of the distribution, respectively. Here the integration of Eq. (1) between zero to infinity is unity. These two parameters for the log-normal distribution can be related to two independent weighted column lengths determined by the X-ray diffraction method assuming certain a grain shape.14

The two weighted average column lengths, area- and volume-weighted average column lengths, were evaluated from the K$_\alpha_1$
components of (111) and (222) peaks using the Fourier method of Warren-Averbach. On (111) and (222) peaks, the Kα2 component was removed by the Rachinger correction\(^\text{(12)}\) and the instrumental peak broadening was corrected by the method of Stokes\(^\text{(12)}\) using a standard specimen of high purity and well annealed Cu mesh. The area- and volume-weighted average column lengths (L(area) and L(vol)) can be determined by plotting normalized Fourier size coefficients against column length.\(^\text{(13)}\)

Assuming spherical grains, we can relate L(area) and L(vol) to \(D_0\) and \(\sigma\) by Eqs. (2) and (3).\(^\text{(13,14)}\)

\[
L(\text{area}) = \frac{2}{3} D_0 \exp\left(\frac{5}{2} \ln^2 \sigma\right) \\
L(\text{vol}) = \frac{3}{4} D_0 \exp\left(\frac{7}{2} \ln^2 \sigma\right)
\]

From the values of L(area) and L(vol) measured for a given specimen, \(D_0\) and \(\sigma\) can be obtained using Eqs. (2) and (3), thus determining the corresponding log-normal distribution function. Example log-normal grain size distribution estimated from the X-ray diffractometry method for the annealed 90 nm wide and 200 nm deep Cu interconnect is shown in Fig. 4.\(^\text{9}\) The log-normal grain size distribution estimated from the X-ray diffraction method is found to be in good agreement with that estimated from the scanning transmission electron microscope (STEM) observation also shown in Fig. 4. Furthermore, it can be said that X-ray diffraction has an advantage for evaluating smaller grain size distributions less than 40 nm.

In Cu interconnects, a significant increase in resistivity has been reported when line widths are less than 100 nm. The resistivity of Cu interconnects with 60–100 nm width and 200 nm depth was reported as a function of mean grain size by Khoo and Onuki.\(^\text{(10)}\) Their result, shown in Fig. 5, indicates that the resistivity of Cu interconnects significantly increases with the decrease of the mean grain size below 70 nm, above which it decreases slightly with the increase of the mean grain size to 105 nm. It is pointed out that the main reason of the increase of resistivity in Cu interconnects with less than 65 nm mean grain size is attributed to the smaller grains at the bottom part of the Cu interconnects. It is, therefore, necessary to clarify the grain size distribution, especially the smaller grain distribution in the bottom part of Cu interconnects.

Figures 6(a)–(c) show the log-normal grain size distributions for the annealed and polished Cu interconnects with electroplated 150, 300 and 450 nm thick overburden Cu films. The log-normal grain size distribution of the bottom half interconnect (the chain curves) is represented taking into account the depth ratio of the whole to bottom half interconnects. For example, the log-normal grain size distribution for the bottom half interconnect shown in Fig. 5(c) is normalized such that the integration of Eq. (1) between zero to infinity is 0.65.

The difference between the grain size distributions of whole and bottom half interconnects corresponds to the grain size distribution
in the top half region of the interconnects. The circles in the figure show the difference between the two distributions at each grain size and the dashed curve represents a least-squares fit of the log-normal function to the difference, i.e., the log-normal grain size distribution of the top half interconnect.

When comparing these three distributions, the crystal grains with relatively small diameter are found to be distributed in the bottom part of these interconnects. This tendency does not change among the Cu interconnects with the three different overburden Cu film thicknesses, i.e., 150, 300 and 450 nm. From longitudinal cross-sectional TEM observation, very small grains in less than 100 nm wide Cu interconnects have been reported to be distributed mainly in the bottom region. These results are in good accord with the present findings. It is clear that the grain size distribution of the bottom half interconnect is little affected by CMP after annealing. Considering the influence of the overburden Cu film, we can see that the crystal grains in the upper part of interconnects near the overburden Cu film grow bigger than in the bottom part of the interconnect, resulting in the grain size distribution in the depth direction of the interconnect. As a whole, grain size becomes bigger with the increase of the overburden Cu film thickness.

Figures 7(a) and (b) plot mean grain size and relative width against the overburden Cu film thickness. In all cases, mean grain sizes in the bottom part are smaller than those in the upper part by about 30%. On the other hand, the relative width becomes smaller with the increase of the overburden Cu film thickness. Also σ is found to be smaller in the upper part than that in the bottom part by about 8% in all cases. In this way, both mean grain size and relative width change, depending on the overburden Cu film thickness. Therefore, it is necessary to evaluate the resistivity of narrow interconnects taking into account of grain size distribution in addition to mean grain size.

The frequency ratio of crystal grains with less than 50 nm diameter in the whole area of interconnects have been used to investigate the resistivity in 30–100 nm wide Cu interconnects. We plots the frequency ratio of crystal grains with less than 40 nm diameter, in which grains are less than mean free path of an electron in Cu, as a function of the overburden Cu film thickness in Fig. 7(c). The frequency ratio in the upper part of the Cu interconnect was quite small and changed little among the Cu interconnects with different overburden Cu film thickness. In the lower part, the frequency ratio was found to be very dependent on the thickness of overburden Cu films in the same way as in the whole interconnect. The frequency ratio is about 20% at the 150 nm thick overburden Cu film, while the frequency ratio at the 450 nm thick overburden Cu film falls to less than 2%.

In Fig. 8, we plotted the frequency ratios of crystal grains with less than 40 nm diameter against mean grain size including those of crystal grains with less than 50 and 60 nm diameter for comparison. Data (closed circles) estimated based on information in previous work on the Cu interconnects with 70–130 nm width, 150–300 nm depth and 160–540 nm overburden Cu film thicknesses are included in this figure. The frequency ratio of crystal grains with less than 60 nm diameter increases almost linearly with the decrease of the mean grain size. The ratio of crystal grains with less than 50 nm diameter decreases also linearly with the decrease of the mean grain size below 80 nm in diameter. The ratio of the crystal grains with less than 40 nm diameter is less than 2% when the mean grain size is larger than 70 nm. This ratio increased gradually with decreasing mean grain size below 70 nm and took value of about 20% at the mean grain size of 55 nm. When comparing Fig. 8 with Fig. 5, the mean grain size dependency of the resistivity in narrow Cu interconnects is nearly the same as that of the frequency ratio of crystal grains with less than 40 nm diameter. Therefore, the resistivity of narrow Cu interconnects is confirmed to strongly depend on the frequency ratio of the crystal grains less than 40 nm in diameter. A large number of crystal grains in interconnects with that high frequency ratio are less than the mean free path of an electron in Cu, which causes an increase in resistivity by electron scattering at the grain boundary. The reduction of the crystal grains with less than 40 nm diameter, which distribute in the bottom part of narrow interconnects, will result in low resistivity interconnects in future ULSIs.
4. Conclusions

We used the X-ray diffraction method to evaluate the crystal grain size distribution in both the upper and bottom part of Cu interconnects with 70 nm width and 150 nm depth as a function of overburden Cu film thickness. In the upper part of the Cu interconnects, the frequency ratio of the crystal grains with less than 40 nm diameter in the whole area of interconnect was found to be quite small and changed little among the Cu interconnects with different overburden Cu film thickness. In the bottom part of the Cu interconnects, the frequency ratio was about 20% at 150 nm thick overburden Cu film, while the frequency ratio decreased to less than 2% at 450 nm thick overburden Cu film. The resistivity of narrow Cu interconnects was found to increase with the increase of the frequency ratio of crystal grains with less than 40 nm, which distribute in the bottom part of narrow interconnects.

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References