Effect of Defects on the Grain and Grain Boundary Strength in Polycrystalline Copper Thin Films

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***Abstract*—In this study, grain boundary quality in terms of order of atomic arrangement of electroplated copper thin films was evaluated by using the IQ (Image Quality) value obtained from an electron back-scatter diffraction (EBSD) method, and the grain and grain boundary strength was evaluated by applying micro tensile test. In addition, in order to investigate the relationship between the strength and grain boundary quality, molecular dynamics (MD) simulations were applied to analyze the deformation behavior of a bicrystal sample and its strength. The variation of the strength and deformation property were attributed to the higher defect density around grain boundaries than that in grains, which impeded the development of slip systems.**

Keywords— Reliability, Electroplated copper, Grain Boundary, Crystallinity, Strength, Molecular dynamics

I. Introduction

Semiconductor devices have been improved in information processing speed by the miniaturization of transistors and improvement of the integration density. Electroplated copper thin films have been used for thin-film interconnections and through-silicon vias (TSVs) in advanced semiconductor devices because of its low electrical resistivity and high thermal conductivity. However, as the diameter of interconnection structures is extraordinarily tiny, it has been getting harder to guarantee the long-term reliability of products because of the various degradation phenomena such as very high local Joule heating and electromigration (EM). It has been reported that EM and SM (stress migration) tends to occur along the grain boundaries in the interconnection material [1,2], and thus, the lifetime of the interconnection components is strongly dominated by the strength of grain boundaries. Electroplated copper thin films often contain porous grain boundaries and the volume ratio of porous grain boundaries in the copper thin films are much larger than that in bulk copper [3]. Since the degradation process of the interconnections is mainly dominated by the self-diffusion of copper atoms along porous grain boundaries, the quality of grain boundaries and its distribution in a thin film should strongly change its physical and chemical properties of the interconnections. In addition to the degradation process caused by the grain boundary diffusion, the high density of porous grain boundaries decreases the thermal conductivity of the thin film interconnection, and some porous grain boundaries with high electrical resistivity cause the localized Joule heating and resulting open failures due to local fusion. Thus, the degradation due to porous grain boundaries becomes particularly prominent in the copper thin film interconnections.

Therefore, it is necessary to quantitatively evaluate the grain and grain boundary strength of electroplated copper films for estimating the lifetime of the interconnection in order to assure the product reliability. In this study, grain boundary quality in terms of order of atomic arrangement was evaluated by using the analysis parameter IQ (Image Quality) value obtained from electron back-scatter diffraction (EBSD) method, and relationship between the strength and quality of electroplated copper thin films was investigated experimentally and theoretically by applying micro tensile test method and molecular dynamics (MD) simulations.

II. Crystallinity-induced Variation of the Strength and Deformation Property

A. *Evaluation Method of the Grain and Grain Boundary Strength Based on the Order of Atom Arrangement*

EBSD analysis is a microstructural crystallographic characterization technique to determine the local crystallographic orientation of grains and quality of grains and grain boundaries based on the order of atom arrangement. The crystallographic quality of grains and grain boundaries of polycrystalline copper thin films was analyzed by using the IQ value [4,5]. In the EBSD method, a small area of a sample (several nm in diameter) is irradiated by a focused electron beam and some inelastic scattered electrons fulfilling the Bragg's diffraction condition produce the so-called Kikuchi pattern consisting of sets of two parallel lines (Kikuchi lines). Since Kikuchi lines are formed by the diffraction of an inelastic scattered electron, the contrast of Kikuchi lines correlates strongly with the intensity of the diffracted beams, and thus, the contrast of Kikuchi lines easily changes depending on the crystallinity, in other words, the order of atom arrangement of the measured area. Therefore, the IQ value indicates the quality of atomic alignment in the measured area. The amplitude of the disorder of the atomic alignment varies depending on the concentrations of various defects such

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as vacancies, dislocations, impurities, and local distribution of strain. In this study, the crystallinity of a grain boundary was evaluated by the IQ value averaged by using the values of the nearby measurement points across the grain boundary [2].

The grain and grain boundary strength was evaluated quantitatively by applying micro tensile test using FIB (Focused Ion Beam) technologies [6, 7]. The micro-tensile test system consists of a test sample, micro-probe, and a both-ends fixed beam of single crystalline silicon as shown in Fig. 1. In this system, the axial and lateral directions of the beam were <110> and <100> crystallographic direction of silicon, respectively. The spring constant of this both ends supported beam is 360 N/m when a load is applied to the center of the beam. Figure 2 shows the SIM (scanning ion microscope) images of set up of this system and micro-scale sample. The sample was fixed rigidly to a silicon beam and a micro probe respectively. The micro probe was pulled up by uniaxial actuator and the deformation of the silicon beam was observed by SIM in real-time until the sample was fractured. The load applied to the sample was calculated by the observed deformation of the beam. In this study, the spatial resolution of the observation was 72 nm. By using these new characterizing methods of materials, it is possible to clarify the relationship between the crystallinity of a grain and a grain boundary and their strength.

The electroplated copper thin films were made by conventional damascene process for evaluating both their crystallinity and strength. Electroplating conditions were as follows. The composition of the plating bath was controlled by diluting 80 g of CuO powder and 186 g of H_2SO_4 into 1000 ml of purified water. The current density during the electroplating of thin film was 10 mA/cm^2. After the electroplating, the fabricated thin film was annealed in pure argon gas at 400°C for 30 min. In order to investigate the relationship between the strength and grain boundary quality of electroplated copper thin films, the bicrystal specimen which had two grains with different crystallographic orientation was cut off from the fabricated thin film by using FIB. Figure 3 shows examples of IPF (inverse pole figure) and IQ maps of the bicrystal specimens. The dash line in this figure indicates the outer frame of the test specimen cut from the thin film. The grain boundary in the center part of the bicrystal specimen was roughly normal to the applied tensile stress. We fabricated four bicrystal specimens with different IQ value of a grain boundary from 5100 to 5900 but almost the same IQ value in grains (about 7000).

B. *Crystallinity-induced Variation of the Strength*

Figures 4 shows the measured stress-strain curves of bicrystal specimens. SIM images at fracture of the specimen with average IQ of a grain boundary 5900 is shown in Fig. 5. The ductile fracture occurred in one grain and no brittle fracture appeared at any grain boundaries in all specimens, indicating that the cleavage strength of a grain boundary was larger than that the critical resolved shear stress (CRSS) of grains. In the micro tensile test, most of the fractures occurred in a grain with higher Schmidt factor, and thus CRSS was calculated for all samples. CRSS is the value of resolved shear stress at which yielding of the grain started to occur, marking

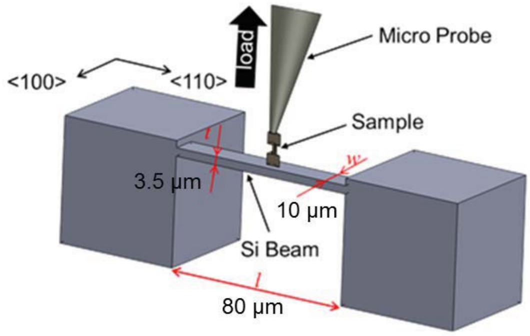

Fig. 1. Schematic image of the micro-tensile system

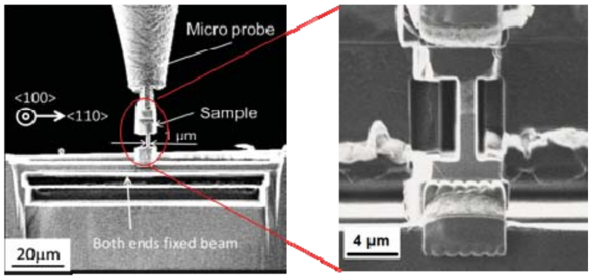

Fig. 2. SIM images of (a) micro-tensile system and (b) micro scale test specimen fixed to a silicon beam and a micro probe

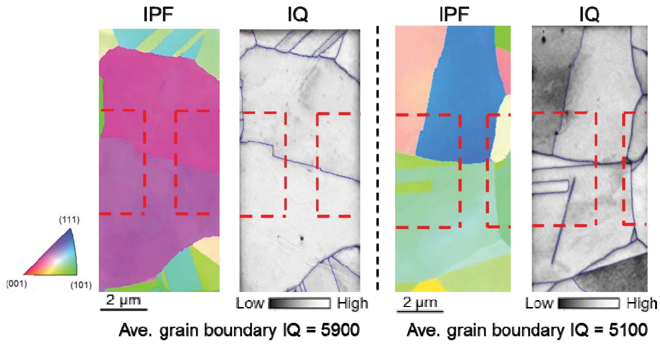

Fig. 3. Examples of IPF and IQ maps of bicrystal specimens. Average grain boundary IQ is average IQ value of measurement points along a grain boundary in the centerprt part of the specimen.

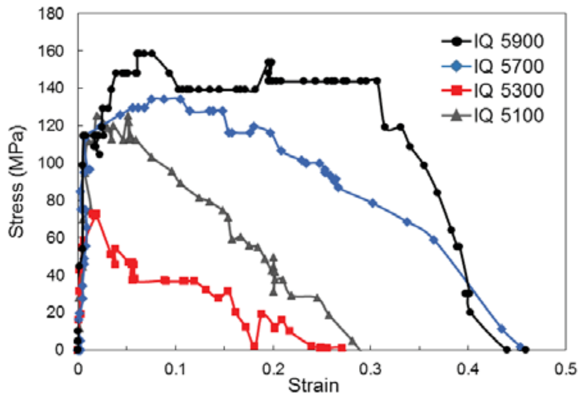

Fig. 4. Stress-strain curves of bicrystal specimens

the onset of plastic deformation. The CRSS and elongation of a copper grain are summarized in Fig. 6 as a function of IQ value of a grain boundary. The elongation decreased monotonically with the decrease of IQ value of a grain boundary, while the difference in the strength was only 19 MPa among four specimens with different IQ value. The almost constant yield strength is considered to be the result of the same crystallinity of grains in all specimens, that is, almost the same density of various defects such as vacancies, dislocations, impurities, and so on that impede the development of slippage in grains.

III. MOLECULAR DYNAMICS SIMULATIONS OF TENSILE TESTS FOR BICRYSTAL STRUCTURE

In order to investigate the relationship between the strength and grain boundary quality, MD simulations were applied to analyze the deformation behavior of a bicrystal structure and its strength. The simulation models were modeled to reproduce the structure of bicrystal specimens (grain boundary IQ = 5900). Based on the EBSD result, the crystallographic orientation of grains in the bicrystal sample and thus, the interfacial crystallographic plane across a grain boundary was defined. In the bicrystal specimen with IQ 5900 shown in Fig, 3, the crystal orientation of upper grain was (hkl)<uvw> = (11-3)<-141>, and that of lower grain was (112)<-110>. In this study, this structure is called (11-3)/(112) model. The contact surface length and width were set around 100 Å respectively, while the tall (vertical length) of each grain was set around 250 Å. Total number of atom was 427,758 atoms. After contacting between the upper grain and the lower grain surfaces, the structural relaxation calculation was performed by changing the volume in order to obtain the equilibrium condition of the bicrystal structure. After this relaxation calculation, it was assumed that obtained structures were under stress (strain) free condition. To investigate the effects of defects on the strength, we considered vacancies by eliminating some atoms randomly from the (11-3)/(112) model. Figure 7 shows the simulation models of copper bicrystal structure with vacancies. The concentration of vacancies which were distributed uniformly in the model was 2 at%, as shown in Fig. 7(b) (vacancy model). The model shown in Fig. 7(c) contained 1 at% vacancies which were only distributed in a range within 25 Å from the grain boundary (grain boundary vacancy model).

The MD simulations were carried out using the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) code [8]. The uni-axial tensile strain was applied by moving the topmost atoms along the z-axis (vertical direction of the grain boundary) from the equilibrium position. The generalized embedded atom method (GEAM) interatomic potential was used and potential parameters of copper were determined to reproduce lattice constant, cohesive energy, elastic constants and melting point. All of the simulations were carried out with 1 fs MD time step at 300 K.

Figure 8 shows the calculated stress-strain curve of the bicrystal models. It was found that the calculated yield stress decreased by introducing vacancies in the structure. On the other hand, the elongation in the grain boundary vacancy model was remarkably small compared with those in no-vacancy and vacancy models. Figure 9 shows the change in the structure during the tensile test. Dislocations and slip bands

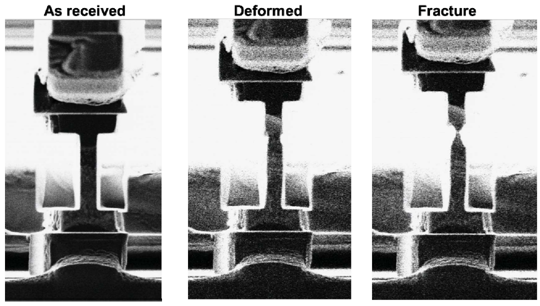

Fig. 5. Defromation behavior and fracture figure of the specimen (IQ=5900)

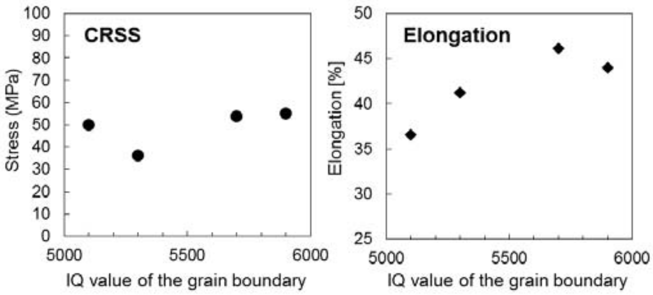

Fig. 6. Relationship between IQ value of the grain boundary and critical resolved shear stress (CRSS), elongation under the micro tensile test

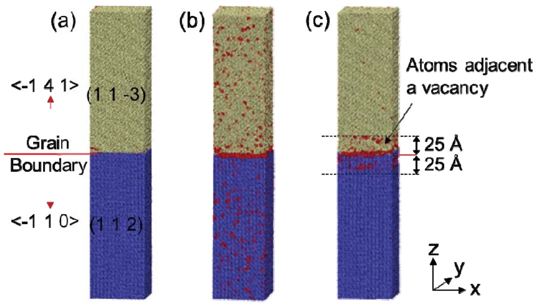

Fig. 7. Simulation models of (11-3)/(112) bicrystal structure

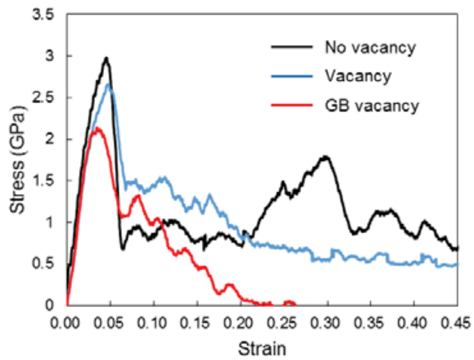

Fig. 8. Calculated stress-strain curve of (11-3)/(112) bicrystal structure

were detected by applying common neighbor analysis (CNA analysis). In this figure, atoms that have hcp structure indicate the stacking faults generated by the slip deformation. Clear plastic deformation occurred in the (11-3)/(112) no-vacancy model. The slippage occurred at (111) plane near the grain boundary in the lower grain firstly. With increasing the tensile stress, the slippage also occurred in the upper grain but the slippage system did not cross the grain boundary in its propagation due to the large amounts of defects around the grain boundary. Finally, the ductile fracture occurred at the grain boundary. This deformation behavior is good agreement with that observed in the experimental micro tensile test shown in Fig. 5.

On the other hand, in the grain boundary vacancy model, small slip occurred around the grain boundary at stress less than yield strength of others. With increasing tensile stress, the slippage was appeared in a localized region around the grain boundary, while the slippage did not occur in the regions outside the high-defect (vacancy) region around the grain boundary. As a result, necking occurred rapidly around the grain boundary and caused final fracture. Since the slippage always initiated in an adjacent crystal near the grain boundary but the concentration of defects around the grain boundary prohibited dislocations from freely propagating on slip planes, the slip system did not develop throughout the grains, which limited the ductility. Therefore, the deformation behavior of bicrystal specimens in which elongation decreased with the decrease of IQ value around grain boundary was attributed to the higher defect density that impedes further propagation of slip initiated near the grain boundary.

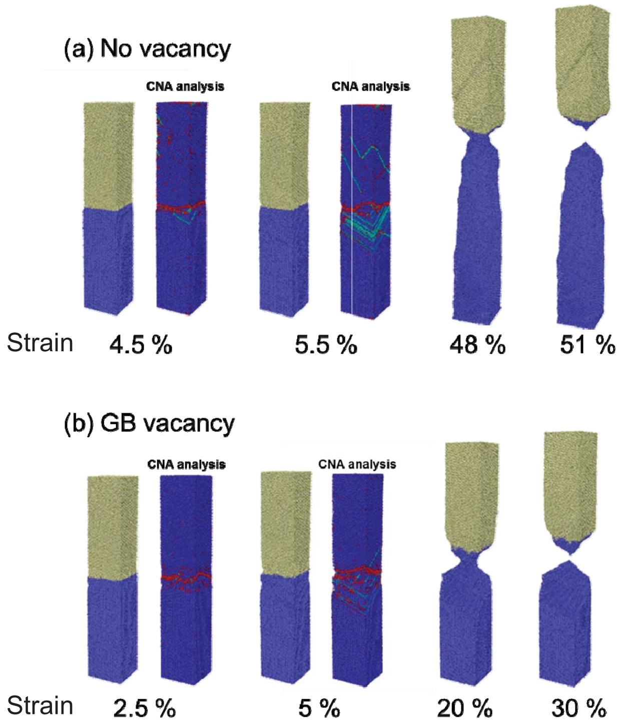

Fig. 9. Deformation behavior of (11-3)/(112) bicrystal structure during the tensile test. In CNA analysis, light blue atoms indicate atoms having hcp structure and red atoms indicate atoms with the number of nearest neighbors less than 12.

IV. CONCLUSION

In this study, relationship between the strength and crystallinity of electroplated copper thin films was investigated experimentally and theoretically. The crystallinity variation of the strength of the bicrystal specimens was evaluated by using both the micro tensile test and EBSD analysis considering the order of atom arrangement. Four kinds of micro scale bicrystal specimens with different crystallinity of a grain boundary were used for this evaluation. The crystallinity of a grain boundary in the specimen was evaluated by using the IQ value obtained from EBSD analysis. The yield strength of the bicrytal specimens was almost constant even when there was a difference in the IQ value of about 1000 between the specimens. On the other hand, the ductility significantly increased with the increase of IQ value. MD simulations showed that the plastic deformation occurred locally around a grain boundary due to the concentration of vacancies around a grain boundary, resulting that the elongation were considerably decreased. The reason for this variation of physical properties between bicrystal specimens was attributed to the higher defect density around a grain boundary than that in grains, which impeded the development of slip systems. It is, therefore, very important to control the crystallinity of the electroplated-copper-base interconnections for assuring the stable and highly reliable operation of semiconductor devices.

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