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Electro- and Thermomigration of Metallic Islands on Si(100) Surface

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Electro- and thermomigration of metallic islands of μm size formed by vapor deposition on the Si(100)2×1 surface have been investigated using an ultrahigh-vacuum scanning electron microscope (UHV-SEM) at substrate temperature higher than the melting point of the islands. The direction of electromigration versus the electric current depends on the type of metal, whereas the direction of thermomigration is always from the cold to hot side independent of the type of metal. The velocity of islands is approximately proportional to the island radius and increases exponentially with temperature for both cases. The activation energies of electro- and thermomigration are approximately 0.6 eV for Au–Si islands having a eutectic composition. The driving forces which act on the islands are discussed based on the diffusion theories for electro- and thermomigration.

KEYWORDS: electromigration, thermomigration, Si surface, ultrahigh-vacuum electron microscopy

1. Introduction

The phenomena of current-induced mass transport, so-called electro-migration, and temperature-induced mass transport, so-called thermomigration, have been investigated for the past 30 years as to impurities in various metals. The most reliable explanations for electro- and thermomigration were given by Hnntington[1)] and Davies,[2)] respectively. However, these phenomena are very complicated, and a number of experimental results have not been explained quantitatively by the theories.

In the present study, it has been found that the metallic islands of μm-order diameter produced on the Si (100) 2×1 surface in ultrahigh vacuum migrate due to the electric current passing through the substrate and the temperature gradient of the substrate surface at temperatures above the melting point of islands. It should be emphasized that electro- and thermomigration occur not only on an atomic scale, but also on a μm scale, such as in islands. The characteristics of island migration due to the electric and thermal effects on the Si surface investigated by means of ultrahigh-vacuum scanning electron microscope (UHV-SEM) are presented in this paper, and the driving forces for island migration are discussed.

2. Experimental

The experiment was carried out using a UHV-SEM (JAMP-30). A p-type silicon wafer with a resistivity of 4–6 Ω cm at room temperature and a size of $20\times5\times0.4$ mm^3 was used as a substrate. The substrate crystal was held between tantalum electrodes and heated by passing a direct current through it, as shown in Fig. 1, for electromigration. On the other hand, thermomigration was observed in the outer portion of the electrodes where there was a temperature gradient between the electrode and the specimen edge. The surface of Si crystals was cleaned by flashing at above 1100°C for a few minutes at a vacuum pressure of less than 5×10^{-10} Torr. The substrate temperature was measured through the resistivity of the substrate, which had been calibrated with an infrared pyrometer. The various metal films of Au, Ni, Pd, Ag, In, Cu, Pt and Al were deposited from heated tungsten wire baskets onto the substrates at room temperature while the thickness was monitored with a quartz thickness microbalance. The evaporation rate was 100 Å/min and the deposited specimens were transferred from the UHV specimen preparation chamber to the UHV-SEM through a transfer tube. The SEM observation was carried out at a primary electron energy of 10 keV.

All metal films were intermixed with the Si substrate at temperatures above 300°C, and islands were nucleated over the surface by further heating at above 500°C, with a Stranski-Krastanov-type growth mode. Compositions of metallic islands were determined through SEM observation by checking the melting points and then referring to phase diagrams of Si-metal binary alloys. From the phase diagrams, we find that Au, Ag and Al islands consist of eutectic alloys, and Ni, Pd, In, Cu and Pt islands are silicides. The migration distance, velocity and direction of migration were measured as functions of substrate temperature, current density and temperature gradient. Dynamic motions of islands for several metal alloys in the inside and outside portions of the electrodes were monitored by TV scan at temperatures above melting points of islands while an electric current flowed through the substrate, and were stored on a video tape.

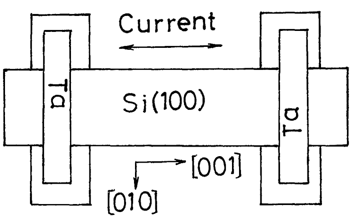

Fig. 1. The schematic diagram of the sample holder. The specimen is held between tantalum electrodes and current direction is along [001].

3. Experimental Results

3.1 *Electromigration*

Electromigration was measured in the middle portion of the specimen, where the temperature was homogeneous. Figure 2(a) is a secondary electron image showing the electromigration for Au–Si islands with passage of the direct current downward, and is a result of electromigration for 10 min at 800°C. The image was taken after cooling the surface to room temperature. The large islands migrate while coalescing with small islands, thus increasing their diameters. The larger the islands are, the faster the speed. By reversing the current, the island migrates along the same route in the opposite direction. However, the velocities are not the same in the two directions, because the increase of the size due to the coalescence with small islands increases the island velocity. It should be pointed out that the direction of electromigration for the Au–Si islands is opposite to that of the electric current.

On the other hand, the Al–Si islands migrate in the direction of the electric current, as shown in Fig. 2(b). Figure 2(b) is the result of electromigration for 10 min at 800°C, and the velocity of Al islands ranged from 0.1 to 0.6 μm/s for micron-size diamter at 800°C. The relationship between velocity and current density for Au–Si islands is shown in Fig. 3 with the parameter of island radius. Figure 3, however, does not show the relationship between the velocity and the current density at constant temperature. Since alternating current is effective only for heating the specimen and does not affect electromigration, we measured the drift velocity of islands while superimposing an alternating current on the direct current to maintain a constant temperature. The experimental results for Pd–Si islands is shown in Fig. 4, which indicates that the velocity of electromigration at constant temperature is proportional to current density.

Similar experiments were carried out for Ni, Pd and Pt islands. The migration direction of these islands was the same as that of Au islands. In further experiments on Ag, In and Cu islands, electromigration was not observed up to the evaporation temperatures. After thermal evaporation of these islands, the surface became

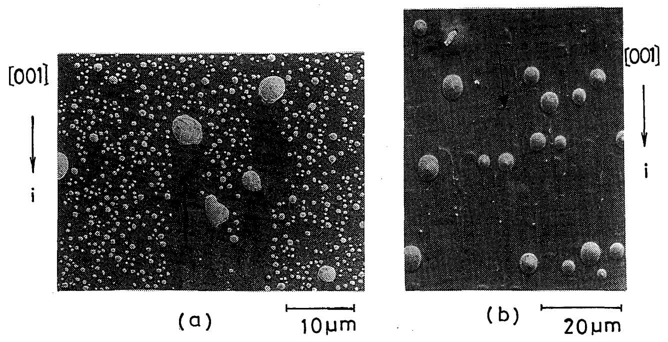

Fig. 2. (a) Electromigration of Au–Si islands and (b) of Al–Si islands for downward direct current. The directions of electromigration for both islands are opposite each other versus the electric current.

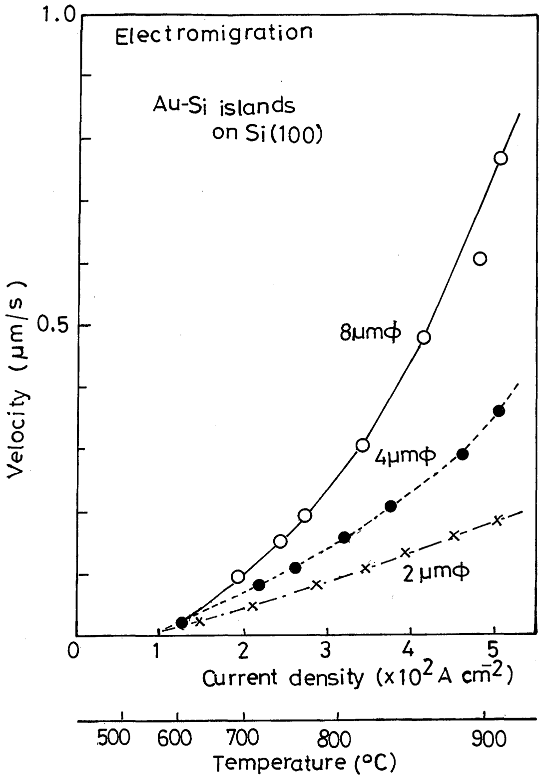

Fig. 3. Relationship between island velocity v and current density j with the parameter of the island radius r. The specimen temperature is a function of j.

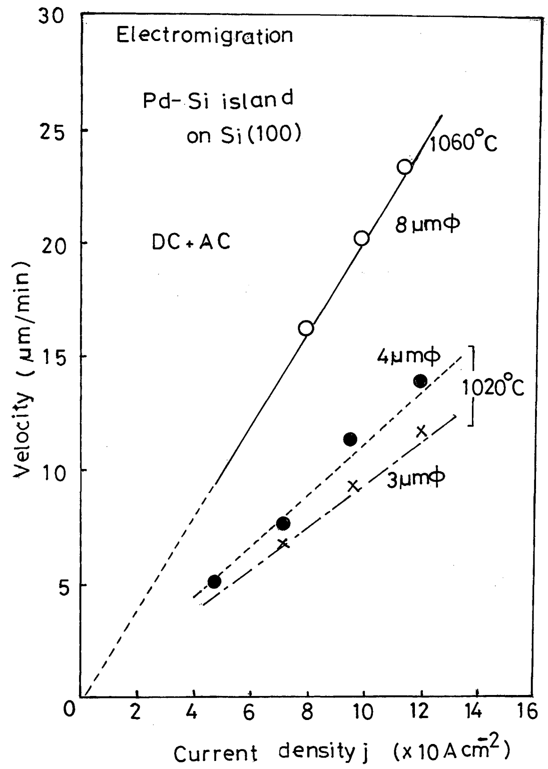

Fig. 4. Relationship between island velocity v and current density j for Pd–Si islands at constant temperature while superimposing alternating current on direct current to maintain a constant temperature. The electromigration velocity is proportional to the current density at constant temperature.

clean, as confirmed with micro-Auger electron spectroscopy.

In addition to these results, we found the following from several trial experiments.

1) For heating with alternating current, no electromigration was observed and the stationary islands were vaporized above the evaporation temperature.

2) The effect of gravity is negligible because the island velocity is not influenced by the inclination angle of the substrate surface up to 90° from the horizontal plane.

3) The velocity of electromigration depends on the crystallographic orientation and is the highest along [110] on the (100) surface.

4) The direction of electromigration is independent of the type of Si-substrate (p- or n-type) with different impurities of concentrations between 10^{-4} and 10^{-6}. Electromigration of Au–Ge islands on a Ge surface was also observed to be in the same direction as that for Si.

5) The trace left after island migration is a clean Si surface and the bottom of the trace is lower than the surrounding substrate surface. The depth of the bottom is 100–600 Å, depending on the type of metal.

3.2 *Thermomigration*

The outside portions of the electrodes with a width of approximately 5 mm, for the Si substrate are not homogeneous in temperature and free from the electric field. The temperature distribution was measured by means of a finely focussed infrared pyrometer with a lateral resolution of 1 mm, and with thermocouples in the vicinities of the electrode and the substrate edge. The temperature distributions as functions of distance from the electrode are shown in Fig. 5 for the heating currents of 3.0 and 3.5 A. The islands grown on the outside portion of the electrode migrate due to the temperature gradient from low to high temperature independent of the variety of metal. Figure 6 shows an example of thermomigration of Au–Si islands in a portion of 500 μm width and between 700 and 800°C. The velocity of the Au–Si islands is 0.1–1.2 μm/s depending on the island radius and the substrate temperature. For thermomigration of islands, it is also found that the larger the islands are, the faster the speed. The relationship between the drift velocity and temperature gradient for islands of 10-μm diameter is shown in Fig. 7 at 750°C. Figure 7 shows that the island velocity is proportional to the temperature gradient.

In addition to thermomigration, we observed island migration due to an electron beam effect. If an electron beam of 0.1-μm diameter with a current density of 10^6 A/cm^2 irradiates a fixed point on the surface at a temperature above the melting point of the islands through heating with alternating current, the islands lying in a circle of 0.1 mm diameter around the electron beam coalesce at its center and form a large liquid island of 10 μm in diameter. Moreover, if the electron beam slowly scans close to a given island, the island moves

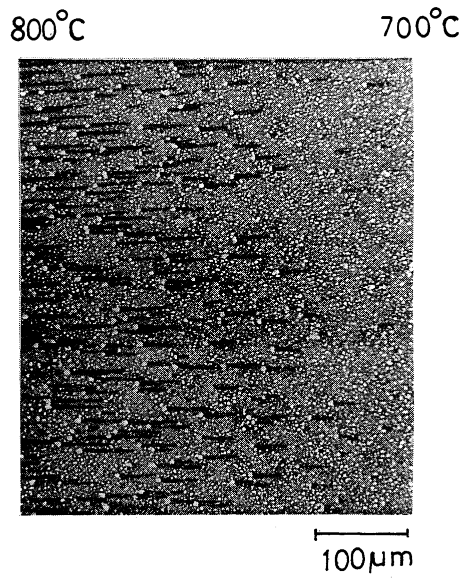

Fig. 6. The thermomigration of Au–Si islands in the outer portion of the electrode in a region of 500-μm width and between 700 and 800°C. The islands drift from low to high temperature.

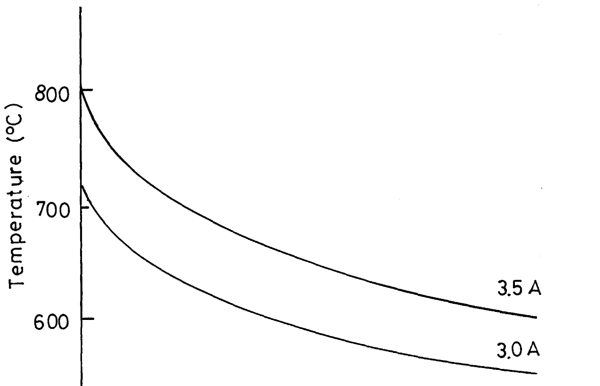

Fig. 5. The temperature distribution of the substrate surface in the outer portion of the electrode measured as a function of the distance from the electrode. The temperatures were measured using a finely focussed infrared pyrometer.

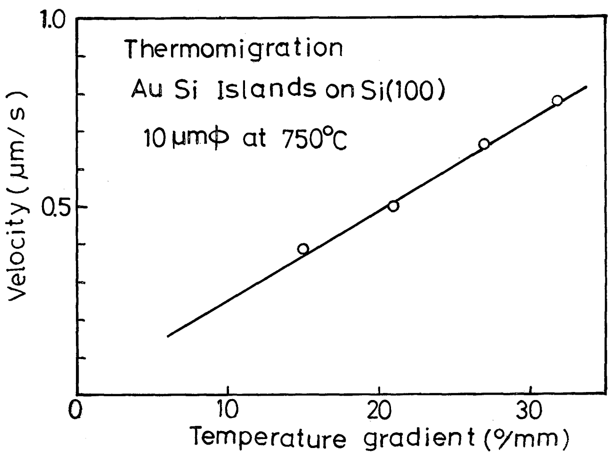

Fig. 7. The thermomigration velocity is proportional to the temperature gradient for Au–Si islands of about 10-μm diameter at substrate temperature of 750°C.

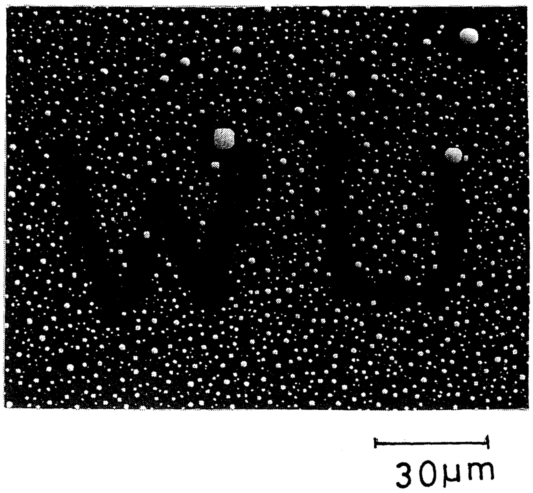

Fig. 8. Letters written using an electron beam on an Au–Si surface at 800°C. A large island lies at the end of each letter.

following the electron beam, as shown in Fig. 8. Thus, we can write letters of micron size by using an electron beam. Figure 8 shows an example of writing the letters "WU" on an Au–Si surface by electron beam scanning. Such an effect is regarded as migration due to the temperature gradient, because every island migrates following the electron beam, independent of the type of metal. Thus, we consider the main effect of an electron beam to be due to the temperature gradient. Here, it should be confirmed that the action of electromigration interacts with thermomigration through the Peltier effect, because the electric current passing through the interface between substrate and island produces a temperature difference at positions between the front and the rear of the islands due to the thermoelectric effect. Therefore, we measured the temperature difference at positions between the front and the rear of a large Au–Si island of 100 μm diameter with the finely focussed infrared pyrometer, while it was migrating with a direct current. No temperature difference in the vicinity of the island was detected within an accuracy of $\pm 5°$ at 800°C. Therefore, we consider both electro- and thermomigration to be independent for islands on the Si surface. Based on the above experimental results, we will discuss the origin of the driving forces of electro- and thermomigration.

4. Discussion

Although the phenomena observed in the present experiment are migrations of liquid islands of μm size, we assume that electro- and thermomigration are caused by the diffusion phenomena of solute atoms against the host Si across the solid-liquid interface due to the electric current and the temperature gradient. The diffusion velocity v of the migrating atoms depending on concentration gradient, the temperature gradient[3-6] and the electric current[7-11] is given as follows, using Fick's diffusion equation:

$$v=-D\frac{\partial \ln c}{\partial x}-\frac{D}{T}\frac{Q^*}{kT}\frac{\partial T}{\partial x}+BeZ^*\rho j, \quad (1)$$

where D is the diffusion coefficient, c is the concentration of solute atoms, Q^* is "apparent heat of transport" for a migrating atom given by Davies,[2] k is Boltzmann's constant, B is mobility, Z^* is the "effective charge" of the migrating atom given by Huntington,[1] ρ is resistivity of a metallic island and j is the current density.

From our experiment, it can be seen that Q^* is negative for every type of metal used in this experiment because all islands migrate from the cold to hot side. The interpretation of thermomigration was described by Jaffe and Shewmon[4] for several impurities in Cu, Au and Ag metals, and it was reported that almost all impurities migrate from the cold to the hot side. The second term in the eq. (1) is expressed by

$$v=-\frac{D_0 Q^*}{kT^2}\frac{\partial T}{\partial x}\exp(-\Delta H/kT). \quad (2)$$

Therefore, Arrhenius plots of $\ln(vT^2)$ versus $1/T$ are obtained, as shown in Fig. 9, as a parameter of $\partial T/\partial x$ for islands of about 10-μm diamter. From Fig. 9, we can obtain the activation energy of 0.6 eV independent of $\partial T/\partial x$.

On the other hand, the third term in eq. (1) is given by

$$v=BeZ^*\rho j=(D_0/kT)\exp(-\Delta H/kT)eZ^*\rho j, \quad (3)$$

where D_0 is a diffusion constant and ΔH is the activation energy for electro-migration. Thus, the usual Arrenius plots of $\ln(vT/j)$ versus $1/T$ shown in Fig. 10 are obtained for three islands of different diameters. From Fig. 10, we can obtain an activation energy of 0.7 eV regardless of the island diameter. Both values estimated from electro- and thermomigration agree within experimental error.

Upon electromigration, there are two sources for the

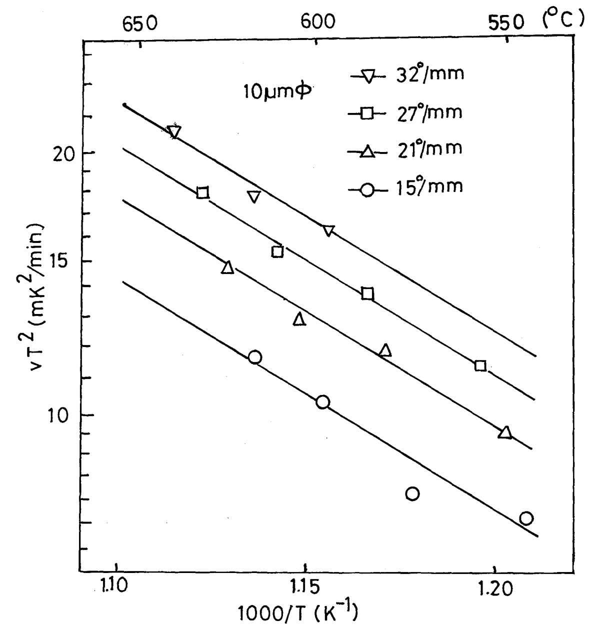

Fig. 9. Arrhenius plots of $\ln(vT^2)$ versus $1/T$ with the parameter of $\partial T/\partial x$ for Au–Si islands of 10-μm diameter. The activation energy is 0.6 eV independent of $\partial T/\partial x$ for thermomigration.

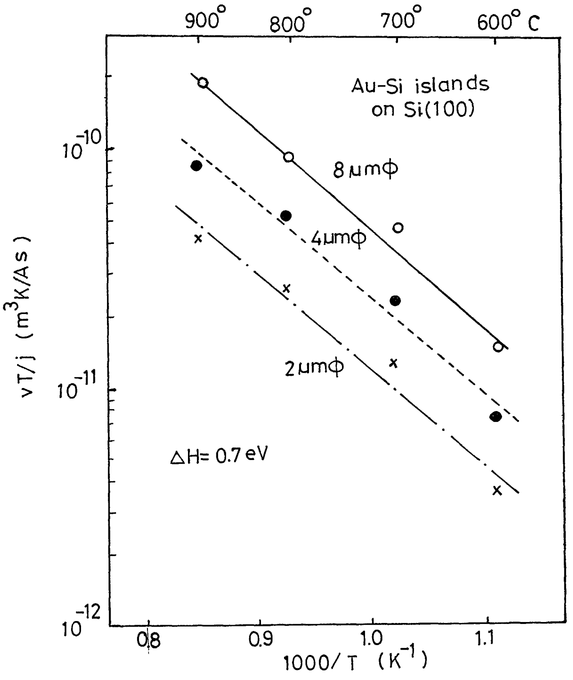

Fig. 10. Arrhenius plots of ln (vT/j) versus $1/T$ for three Au–Si islands of different diameters. The activation energy is 0.7 eV independent of their diameters for electromigration.

driving force: the first arises from the direct action of the external field on the charge of the migrating ion ("direct force") and the second from the scattering of conduction electrons by solute atoms ("wind force"). Verbruggen[7] demonstrated that the driving force $\boldsymbol{F}$ for electromigration is given by an effective charge Z^* of the solute atoms under the existence of a macroscopic electric field $\boldsymbol{E}$.

$$\boldsymbol{F}=(Z+Z_{\text{wind}})e\boldsymbol{E}=Z^*e\boldsymbol{E}. \quad (4)$$

Here, Z and Z_{wind} are, respectively, a bare charge of the migrating atom and an effective charge corresponding to the wind force. The direct force is independent of variation of the current density j, whereas the wind force is proportional to j. Therefore, Z and Z_{wind}, which are components of Z^*, can be separated, in principle, by the measurement of ρ with varying specimen temperature at a constant electric field. However, the magnitude and sign of Z and Z_{wind} obtained from the experiment for the group-V transition metals were not explainable theoretically.[7] This is a result of a flaw in the theories for calculation of the nature of the screening of the electric field at the site of an impurity by conduction electrons and the existence of inhomogeneities in the electric field and current flow near an impurity.

In semiconductors, however, the "direct force" is of great importance because the resistivity of a semiconductor is greater than that of metal. Therefore, the charge transferred from the host Si to the migrating atom, which is probably deduced from a relative value of electronegativity or work function between metal and silicon, is a rough indicator for estimating electromigration direction. In fact, opposite directions of electromigration between Au and Al are interpretated as the electronegativity of Au being larger than that of Si, and of Al being smaller than that of Si.[12]

The driving forces generated by the concentration gradient, the temperature gradient and the electric field, F_c, F_T and F_E, are given by $kT\nabla \ln c$, $Q^*\nabla \ln T$ and eZ^*E, respectively. Taking $\nabla \ln c=30\ \text{m}^{-1}$, $Q^*=-8$ kcal/mol (from ref. 4), $\nabla \ln T=300\ \text{m}^{-1}$, $Z^*=1$ and $E=400$ V/m, $F_c=4\times10^{-19}$ N/atom, $F_T=1\times10^{-17}$ N/atom and $F_E=6\times10^{-17}$ N/atom. Since gravity acting on a migrating atom is on the order of 5×10^{-21} N/atom, the forces of electro- and thermomigration are much larger than those of the concentration gradient and gravity. The total force acting on an island is the sum of the force acting on the individual metal atoms contained in an effective volume sinking below the substrate surface. Hence the forces of electro- and thermomigration for an island of 1-μm diamter are on the order of 10^{-9}–10^{-10} N, which are larger than gravity or the force due to the concentration gradient, 10^{-12} N.

Furthermore, it should be noted that the island velocities of electro- and thermomigration measured in the present experiment are three orders higher than that of impurities in metals compared with refs. 4 and 9. This is due to a large difference in diffusion coefficients between metal and semiconductor. In fact, the diffusion coefficients at 800°C for several impurities in metal are on the order of 10^{-9} cm²/s which is smaller than those in Si, 10^{-5} cm²/s. This is caused by a difference in the diffusion mechanisms between metal and semiconductor. The values of Z^* and Q^* are probably of the same order for impurities in metal and semiconductor.

The size dependence of the islands for electro- and thermomigration may be explained as follows: the driving force is probably proportional to r^2 (r is island radius), because the effective volume acting on an island for electro- and thermomigration is that of the bottom sinking below the substrate surface surrounding the island. On the other hand, the island velocity is nearly constant at a given temperature, hence the total driving force acting on an island should be balanced with the reaction force which is probably a horizontal component of the surface tension proportional to the island radius. Thus, the velocity of islands for electro- and thermomigration is proportional to the island radius.

The interpretation in this paper is one of the hypotheses assuming impurity diffusion of individual metal atoms across the liquid-solid interface. A more exact theory of these phenomena will be presented in the near future.

5. Conclusions

In conclusion, *in situ* observations using the UHV-SEM enable us to demonstrate how electro- and thermomigration of metallic islands of μm size proceed on a Si surface as a result of electric and thermal effects. Such phenomena involving large-scale islands were observed for the first time in this experiment, and are of great interest in the field of mass transport due to the electric field and thermal gradient, and for application in the semiconductor industry for understanding the process of electro- and thermomigration in integrated circuits. The authors will report more quantitative results elsewhere for metallic islands other than Au–Si.

Acknowledgment

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