SURFACE MORPHOLOGIES OF ALUMINUM FILMS ON SILICONE OIL SURFACES*

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Abstract: The surface morphology and growth mechanism of an aluminum film system deposited on silicone oil surfaces by a vapor depositing method was investigated by scanning electron microscopy. It was found that the perpendicular fluctuation of the film's bottom surface was more remarkable than that of the film's top surface. Near the joint between the film on the silicone oil substrate and the film on the silicon wafer surface on which the silicone oil substrate rested, was a naturally formed anomalous wedge-shaped wrinkly structure with slopes of $10^{-4} - 10^{-5}$ rad, whose growth mechanism could be interpreted under the assumption of the thermal expansion behavior of the liquid substrates.

Key words: surface morphology, growth mechanism, liquid substrate, aluminum(Al)
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INTRODUCTION

The nature of substrates has a crucial effect on both the microstructure of the films and the growth mechanism (Haus et al., 1987; Krug et al., 1990). For instance, the top surfaces of metallic thin films deposited on solid substrates generally have a rough structure, which can be described by the self-affine characteristics (Palasantzas et al., 1994). For thick films, it is not necessary to consider the roughness of the upper surfaces since it will not influence obviously any physical properties of the films. For very thin films, however, since the perpendicular fluctuation of the upper surfaces is near the order of the film thickness, the rough surface effect becomes very strong and should not be disregarded (Ye et al., 1995; Ye et al., 1996).

To our best knowledge, the morphology of the film's bottom surface in contact with the solid substrate, has rarely been studied experimentally so far. The main reasons are: (1) It is difficult to entirely remove the film from the solid substrate since they are tightly connected to each other. Although several techniques had been developed to resolve this problem (Julien et al., 1992; Meakin et al., 1993), they could not be used for studying the bottom surface directly since, in these techniques, other materials (soluble painted layers, for instance) are usually introduced between the films and the substrates. (2) It is naturally believed that there is no difference between the height-contour plots of the bottom surface and the solid substrate surface since the film grows originally on the substrate. It should be noted that, however, this is not always true, since various defects (holes, grain boundaries, etc.) may form on the bottom surfaces. Therefore, below submicron length scales, the bottom surface of the film, the surface of the substrate and the interface between the film and substrate are three different elements.

In this work, we study the surface morphology and growth mechanism of a continuous Al film system deposited on silicone oil surfaces. We will show that, in the film thickness range $d = 10 - 60$ nm, the top and bottom surfaces exhibit a characteristic granular structure and that the average granular size of the top surface is obviously smaller than that of the bottom surface. During the deposition, an anomalous wedge-

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shaped structure near the edges of the films naturally formed. Our experimental result indicates that this anomalous structure results from the movable behavior of the liquid substrate.

EXPERIMENTAL DETAILS

The film samples were prepared by thermal evaporation of 99.999 % pure AI at pressure of 5×10^{-4} Pa and room temperature. A small pure silicone oil (Dow Corning 705 Diffusion Pump Fluid with a vapor pressure below 10^{-8} Pa at room temperature) drop with diameter $\phi \approx 1.2$ mm was dripped on a piece of polished silicon wafer surface [Si(111)] (We use silicon wafers here since it is quite helpful for the SEM measurement. In fact, our experiment indicated that the results reported in this paper will not change obviously if we use glass substrates rather than the silicon wafers). Thus a liquid substrate with surface area of about 3 mm^2 was formed over the solid substrate. The silicon wafer was horizontally fixed 200 mm above the filament (tungsten). The deposition rate and the film thickness were controlled by a quartz-crystal thickness monitor located just beside the substrate.

In order to characterize the top and bottom surfaces of the films, a special technique was developed to separate the Al films from the oil substrates. This technique included two steps: (1) After the deposition process, the sample was removed from the vacuum chamber. Then a clean polished silicon wafer [Si(111)] was carefully made to touch the Al film's top surface, which would stick immediately on the silicon wafer surface. After washing with acetone, the clean bottom surface of the film would appear on the silicon wafer surface. (2) In order to image the top surface of the Al film, a small silicone oil drop was first dripped onto an about 3 mm^2 area frosted glass surface used to carefully contact the top surface of the Al film deposited on the oil substrate and then separate from the sample again. Fortunately, at the moment, the Al film broke into many small pieces, with some of them sticking on the oil drop dripped on the frosted glass surface. It should be noted that, on this oil drop, the film surface that faced to us was the bottom surface. We found that no oil stuck on the bottom surface at this stage since the oil did not wet the Al film. After Step (1) described above was repeated, a clean top surface of the Al film appeared on the silicon wafer surface. Scanning electron microscopy (SEM) measurement was then taken immediately under atmospheric condition.

The new and original technique described above is quite simple and effective for detailed study of a film's top and bottom surfaces.

RESULTS AND DISCUSSION

Figs.1(a) and (b) present the typical SEM images of the top and bottom surfaces of a sample, and show that metallic films can be fabricated on liquid substrates. Fig.1(c) shows the morphology of the top surface of an Al film on a polished silicon [Si(111)] wafer surface. Figs. 1(a) and (b) show similar granular structure at this length scale, but with the top surface being more concentrated and more smooth. The top surface's vertical fluctuation and average granu-

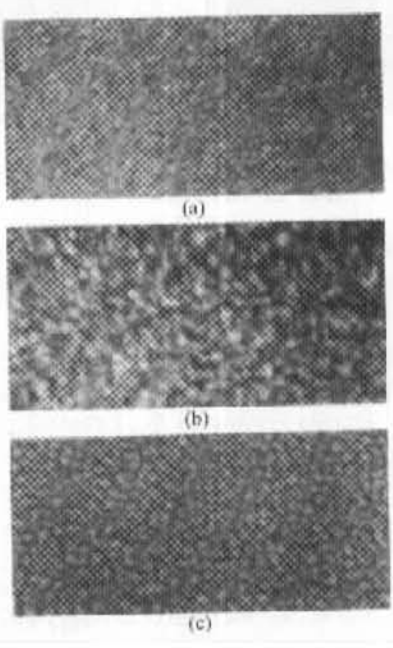

Fig.1 Typical SEM images of Al film

(a) the top surface of the film deposited on silicone oil surface;

(b) the bottom surface of the film deposited on silicone oil surface;

(c) SEM image of the top surface of Al film deposited on a polished silicon wafer surface($d = 30.0$ nm, $f = 0.05$ nm/s, each image area 1.0×2.1 μm^2)

lar size were both obviously smaller than those of the bottom surface. Compared with the Al film on the solid substrate (Fig. 1c), the surfaces of the film deposited on the liquid substrate were rougher and less uniform. We believe that with increase of the film thickness, the liquid surface effect gradually weakens and the top surface morphology of the film on the liquid surface will approach that of the films on solid substrates. However, further theoretical study on its mechanism is still needed.

The most exciting and unexpected result (Fig. 2) was the existence of an anomalous wedge-shaped wrinkly structure near the edge of the film's bottom surface. The average length of the wrinkles period was about $10^{-1} - 10$ μm. The contour lines of the wrinkles around the oil drop could be the key to the formation mechanism of the wrinkly structure.

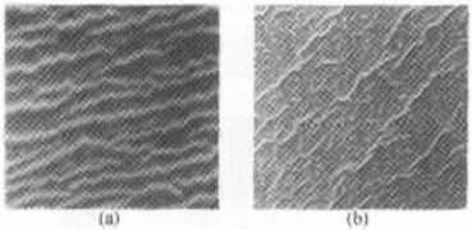

Fig. 2 SEM images of the wrinkly structures near the edges of the film's bottom surface $d = 30.0$ nm

(a) $f = 0.05$ nm/s, image area 6×6 μm^2;
(b) $f = 0.30$ nm/s, image area 20×20 μm^2

Detailed observation showed that the oil drop expanded steadily during the deposition. After the Al film was separated from the oil drop, the residual oil on the solid substrate, i.e., the polished silicon wafer surface, was then cleaned with acetone. Fig. 3 of the Al films deposited on the silicon wafer substrates shows a ring on each silicon wafer sample's surface: the inner and outer circles of the ring are the respective marks of the oil drop before and after the deposition, which showed obviously that there was no film in the area of the inner circle since it was always covered with the oil during the deposition; and that the thickness of the Al film in the area outside the ring equaled the nominal thickness of the film on the middle part of the oil drop since it was always uncovered. Optical microscope observation showed that the thickness of the film on the silicon wafer surface increased almost linearly from the inner radius r_1 to the outer radius r_2 of the ring, indicating that radius r of the oil drop increased uniformly during the deposition. According to the values of r_1, r_2 and the nominal film thickness d, we concluded that the slopes of the wedge-shaped rings in Figs. 3(a) and 3(b) are about 10^{-5} rad and 10^{-4} rad respectively. We concluded from Fig. 3 that for a fixed film thickness, the total increment of the oil radius, i.e., $\Delta r = r_2 - r_1$, decreases obviously with the increase of the deposition rate f. Furthermore, we found that for fixed deposition rate and nominal film thickness, the increment Δr was nearly proportional to r_1 in the range $r_1 = 0.4 - 2.0$ mm.

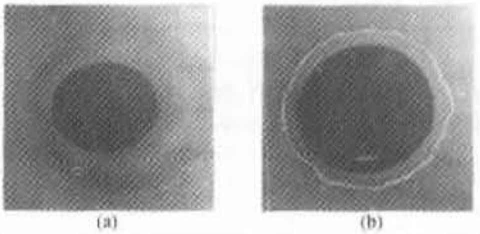

Fig. 3 The full view of the Al films on the silicone wafer surfaces. The inner and outer circles are the marks of the oil drops before and after the deposition respectively $d = 30.0$ nm

(a) $f = 0.05$ nm/s, image area 2.7×2.7 mm^2;
(b) $f = 0.30$ nm/s, image area 2×2 mm^2

Our thermocouple showed that during the deposition, the total temperature increment of the silicon wafer surface was around 0.5 ℃ and changed with the tungsten filament temperature and the deposition time. Therefore, we believe that this expansive phenomenon resulted from the considerable rise in the local temperature of the oil drop surface during the deposition because of the heat radiation from the tungsten filament and bombardment by Al atoms.

According to the experimental phenomenon above, we therefore propose that the wrinkly structure near the edge of the Al film results from the movable behavior of the liquid substrate. During the deposition, after the oil drop had been covered with some layers of Al atoms, the volume of the oil drop increased and its surface tension decreased due to the considerable rise in the local temperature. Then the oil broke the connection between the Al film on the oil surface and the Al film on the silicon wafer surface. At this moment, the film in the shape of a disc floated on the oil drop surface with diameter slightly smaller than that of the oil drop. However, the oil surrounding the Al disc was covered soon by the Al atoms since the thermal deposition was in progress. Therefore, a step surrounding the edge of the film disc was formed. This process continued and finally the whole wedge-shaped films on both the oil substrate and the silicon wafer surface were formed, as shown in Fig.4. As discussed above, the slope of the wedge-shaped edge of the film on the oil surface should be close to that of the Al film on the silicon wafer surface (Fig.3). Therefore, since the film on the oil drop was approximately a freely standing film without internal tension, then wrinkles formed along the contours surrounding the edge of the film (Fig.2) since a corrugated film is often a lower energy system than a flat film system (Williams et al., 1995). However, this wrinkling phenomenon will not occur in the wedge-shaped film on the silicon wafer surface because of the strong interaction between the Al film and the solid substrates.

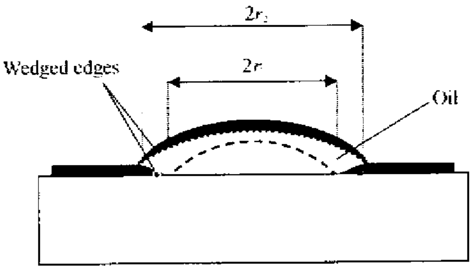

Fig.4 Schematic representation of the growth mechanism of the Al films (cross section). The shaded parts are the Al films. The dash and dot curves denote the outlines of the oil drop before and after the deposition respectively

Furthermore, our experiment showed that in the film thickness range $d = 10 - 60$ nm, Al films deposited on the oil surfaces showed similar structures as described in Figs. 2 – 4. We believe that the phenomenon described in Fig.4 can be used to fabricate various metallic rough films for various purposes. The lens-shaped film on the oil surface and the aperture-shaped film on the silicon wafer surface shown in Fig.4 are good examples in this new field.

CONCLUSIONS

In conclusion, we have described a method for preparing an Al film system on liquid substrate. The Al films were removed from the liquid substrates successfully and different granular morphologies of the top and bottom surfaces were observed. Near the edges of the films on both the oil and the silicon wafer surfaces, a characteristic wedge-shaped structure formed. We show that this structure resulted from the thermal expansion property of the liquid substrate and therefore the slope of the wedge-shaped edge can be controlled easily in experiments. The anomalous wrinkles can be explained by assuming that, for a freely standing sample, a corrugated film is often a lower energy system than that of a flat one. We believe that this movable substrate deposition method will present us a new way for preparing other new kinds of rough film systems.

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