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ANOMALOUS HOPPING AND TUNNELING EFFECTS IN A NEW ALUMINUM FILM PERCOLATION SYSTEM*

XIA A-gen(夏阿根), YE Jian-ting(叶剑挺), YE Quan-lin(叶全林),
TAO Xiang-ming(陶向明), JIN Jin-sheng(金进生), YE Gao-xiang(叶高翔)

(*Department of Physics, Zhejiang University, Hangzhou 310028, China*)

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Abstract: A new aluminum thin film percolation system, deposited on glass and silicon wafer surfaces by a vapor deposition method, was investigated. By using the expansive and mobile behaviors of the silicone oil, the Al films are quenched gradually by the silicone oil during the deposition process. The *R-I* behavior of the film system was studied, and the anomalous conductivity indicated that, at very low current, the hopping and tunneling effects in the films are much stronger than those of the normal film systems.

Key words: thin film, percolation, hopping conductance, tunneling effect
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INTRODUCTION

The microstructure of thin films plays a crucial role in a wide variety of physical processes. There are clear evidences that many physical properties of a thin film vary dramatically as its microstructure changes (Haus et al., 1987). For instance, rough substrates can be used to fabricate a bilateral rough film system with surface morphology and its electrical behaviors are much different from those of the flat systems (Ye et al., 1994a; 1994b). So it is expected that the microstructure of films can be well controlled or designed in more effective ways so that the films exhibit desirable properties.

Ye et al. reported an experiment on silver atom deposition on silicone oil surfaces by vapor deposition method (Ye et al., 1996; Ye et al., 1998a 1998b; Yang et al., 1999). The metallic thin films formed on the liquid substrates exhibited characteristic percolation structure and anomalous values of the third-harmonic coefficients and the current breakdown exponent, indicating that the physical mechanism of the electrical breakdown process in this system is quite different from that of the percolation films deposited on solid substrates. The result above presents us an example that like the solid substrate, liquid surfaces can also be used as film substrates. Therefore, it is expected that more new film systems can be prepared by using the mobile behavior of the liquid substrates.

The microgeometry of a percolation film can be considered as a random resistance network with total sheet resistance R, carrying a current I, made up of elements of resistance r_α carrying currents i_α. Theoretical analysis of this random resistor network (RRN) yielded the following model (Ye et al., 1994b)

$$B = R_0^{2+w} \qquad (1)$$

where R_0 is the zero-power sheet resistance, w is a critical exponent and B is the normalized third-harmonic coefficient, which is related to the resistance fluctuation. The coefficient B_0, which equals the value of B when the frequency of the current approaches zero, is obtained from the dc R-I relation of the films (Ye et al., 1994b)

$$R = R_0 + B_0 I^2 \qquad (2)$$

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It should be noted that this quadratic *R-I* behavior is interpreted in terms of the rise in the temperature of the hot spots (or links) in the films due to the local Joule heating and the microstructure of the percolation film remains unchanged.

In this paper, we report the results on the growth mechanism and the nonlinear *R-I* behavior of a new film percolation system deposited on silicon wafer and glass surfaces. It was observed that, at very low currents, the sheet resistivity decreased dramatically with the current, indicating that there are strong hopping and tunneling effects in the samples due to the characteristic microstructure of the films.

EXPERIMENTAL DETAILS

The samples were prepared by thermal evaporation of 99.999 % pure aluminum at pressure of 6×10^{-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 $\phi \approx 2 - 4$ mm was dripped on a piece of glass or silicon wafer surfaces. The oil drop here was used to quench the Al film during the deposition (see the description below). The filament (tungsten) - substrate distance was about 200 mm. The deposition rate f and the film thickness d were controlled by a quartz-crystal thickness monitor located just beside the substrate. After the deposition, the films were removed from the vacuum chamber and the dc sheet resistance R of the samples as a function of the dc current I was then measured in both air and vacuum (pressure $p < 1 \times 10^{-3}$ Pa) conditions with the four-probe method.

RESULTS AND DISCUSSION

Microstructure and growth mechanism

After the samples were ready, the oil drops on the silicon wafer (or glass) surfaces as well as the Al films on the oil surfaces were washed cleanly with acetone. The full view of the Al films on the silicon wafer substrates is shown in Fig. 1 showing that, for each sample, there is a ring on the silicon wafer surface: the inner and outer circles of the ring are the marks of the oil drop before and after the deposition, respectively. The phenomenon shown in Fig. 1 suggests that the oil drops expanded during the deposition. Obviously, there is no Al atoms within the area of the inner circle since it was always covered with the oil during the deposition, and the thickness of the Al film in the area outside the ring equals the nominal thickness detected by the quartz-crystal thickness monitor since it was always uncovered. With an optical microscope, we find that, approximately, the thickness of the Al films on the silicon wafer (or glass) surfaces increases linearly from the inner radius r_1 to the outer radius r_2 of the ring, indicating that, approximately, the radius r of the oil drop increases uniformly during the deposition. According to the values of r_1, r_2 and the nominal film thickness d, we get that the slope of the quasi-wedge ring is given by $\theta = d/(r_2 - r_1)$ approximately. Therefore, in Figs. 1(a) and (b), we find $\theta \approx 10^{-5}$ rad and 10^{-4} rad, respectively. One can conclude from Fig. 1 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, for a fixed deposition rate ($f = 0.05$ nm/sec) and nominal film thickness ($d = 30$ nm), we find that the increment Δr increases linearly with the diameter $2r_1$ and that the slope of the fit line equals 0.12 ± 0.01, as shown in Fig. 2, indicating the expansion is related to the total

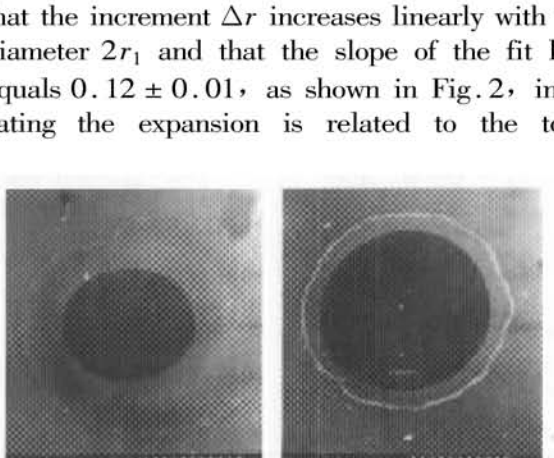

Fig. 1 Full view of the Al films on the silicon wafer surfaces. The inner and outer circles are the marks of the oil drops before and after the deposition, respectively. $d = 30.0$ nm. (a) $f = 0.05$ nm/s, image area is 2.7×2.7 mm^2; (b) $f = 0.30$ nm/s, image area is 2×2 mm^2.

amount of the oil. These phenomena above provide a method for us to effectively control the slope of the quasi-wedge film in experiment.

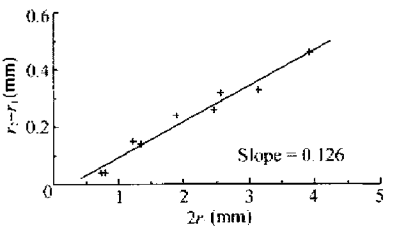

Fig.2 The total expansion distance Δr as a function of the diameter $2r_1$.

Our thermocouple detected in experiment that, during the deposition, the total temperature increment of the silicon wafer surface was around 0.5 ℃ and changed with the filament temperature and the deposition time. Therefore, we believe that the expansive phenomenon described above is resulted from the considerable rise in the local temperature of the oil drop surface during the deposition because of both the heat radiation from the filament and strike by the Al atoms.

According to the experimental phenomena above, we therefore propose that, during the deposition, when the silicon wafer (or glass) surface near and around the oil drop has been covered with some layers of Al atoms, the volume of the oil drop increases and its surface tension decreases due to the considerable rise in the local temperature. Then the oil expands and the diameter of the oil drop increases. Therefore, the Al atom layers near the oil drop will be covered gradually with the oil. This process continues and finally a new metallic quasi-wedge film forms, as shown in Fig. 1. When the Al atom layers are covered with oil, the kinetic energy of the Al atoms will be dissipated and the thermal diffusion of the Al atoms will be blocked by the oil molecules. Thus, a huge number of defects and a characteristic percolation structure forms in the Al film due to the immediate quenching process.

The growth mechanism described above can be summarized in Fig. 3. We believe that the mechanism in Fig.3 can also be used to fabricate other new metallic rough films for different purposes. The aperture-shaped film on the silicon wafer surface shown in Figs. 1 and 3 are good examples in this new field.

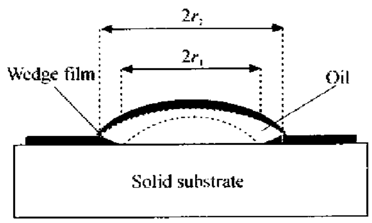

Fig. 3 Schematic representation of the growth mechanism of the Al films (cross section). The shaded parts are the Al films. The dash and dot curves denote the outlines of the oil drop before and after the deposition, respectively.

Anomalous hopping and tunneling effects

If the above qualitative model of the growth mechanism is correct, we expect that the microstructure of the samples is characteristic and therefore the dc conductivity behavior of the samples will be quite different from those of the other systems.

The dc current dependence of the sheet resistivity of the new quasi-wedge ring films was systematically measured at room temperature, as shown in Figs.4 and 5. The slope of the quasi-wedge film sample is of the order of $\theta = 10^{-5}$ rad, the maximum thickness of the film is d = 20 nm, and the increment Δr of the film is 10 mm (In this experiment, in order to increase the increment Δr in a desirable direction, the glass substrate was fixed slightly deviating from the horizontal position). Then this quasi-wedge ring film deposited on glass surface was carefully carved and shaped with a small knife so that a band-like film of width w was obtained. If the thickness d' of the film changes linearly as described above, then we obtain $d' \approx \frac{d}{r_2 - r_1}(r - r_1)$. Therefore the thickness of the band-like film can be selected by choosing the different area of the quasi-wedge film to shape the band-like film. In our experiment, since the width w was very narrow ($w \approx 0.5$ mm) and the films were carved along the circumference of the rings, the maximum difference of the thickness d' in different areas of each band-like film in Fig. 4 was less than 0.5 nm, which can therefore be considered approximately as homogeneous films. The size of each film was $2.9 \times 0.5\ mm^2$. The dc *R-I* behavior of the band-like films is shown in Fig. 4.

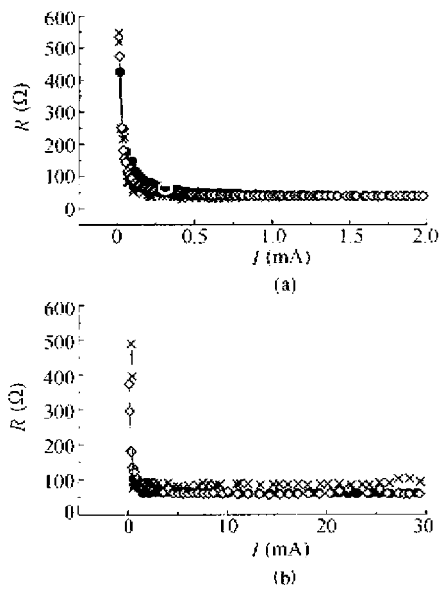

Fig. 4 ***R-I*** **characteristics of band-like Al films. The band-like films were carved from the wedge-shaped film with $\theta = 10^{-5}$ rad, $d = 20$ nm and $\Delta r = 10$ mm. $f = 0.005$ nm/s. (•) and (•) denote the first and second measurements in vacuum respectively; (×) denotes the third measurement in the air. (a) $d' = 4.0 \pm 1.0$ nm; (b) $d' = 15.0 \pm 1.0$ nm.**

Within our measurement ranges, one can see from Fig. 4 that the sheet resistance drops quickly with the current at very low currents, and that the absolute values of the slopes, i.e., $\left|\frac{dR}{dI}\right|$ in Figs. 4(a) and (b) are around 700 and 0.5 Ω/mA respectively, which are anomalous behaviors compared with those of the traditional film systems (Ye et al., 1994b; Yagil et al., 1992; Song et al., 1992; Ye et al., 1995; Ye et al., 1996). In Fig. 4, $\frac{dR}{dI}$ approaches zero and approaches staturation with further increase of the current. Then the sheet resistance increases slowly with the current, which results in a positive value of $\frac{dR}{dI}$, as shown in Fig. 5. Finally, a large resistance fluctuation appears and the microstructure of the films is damaged by the Joule heating. On the other hand, we see from Fig. 4 that both R and $\frac{dR}{dI}$ remain almost unchanged after the first measurement and furthermore there is approximately no difference between the result measured in air and that in vacuum condition. Therefore, the *R-I* behavior in Fig. 4 is approximately a reversible behavior and corresponds to the microstructure of the films.

We propose that the nonlinear *R-I* behaviors described in Figs. 4 and 5 mainly resulted from the characteristic microstructure of the films since other systems do not exhibit such behaviors (Ye et al., 1994b; Yagil et al., 1992; Song et al., 1992; Ye et al., 1995; Ye et al., 1996). Since the film, like various metal-insulator-metal (MIM) tunneling diodes, contains many defects and has a characteristic percolation structure, many weak links are constructed in the films. When the current passes through a weak link, the local temperature change is sufficient to excite local hopping and tunneling effects. Both effects would reduce the current density of the link and therefore the sheet resistance. The higher the current is, the more the amount of the resistance will be reduced, which causes weak third-harmonic signals. Therefore, the phenomenon of $\frac{dR}{dI} < 0$ is mainly due to the short circuit due to the hopping and tunneling effects. When all the weak MIM junctions have been broken down, the hopping and tunneling processes become saturated and then the change of the sheet resistance is mainly resulted from the Joule heating effect, which will result in a positive slope, i.e., > 0 (Fig. 5).

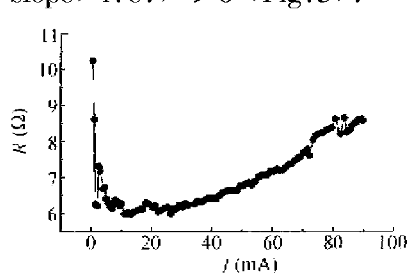

Fig. 5 **Full view of the *R-I* characteristics of the sample in Fig. 4(b) (the dot curve).**

We note that, at very low currents, the value of $\left|\frac{dR}{dI}\right|$ in Fig. 4(a) is much larger than that in Fig. 4(b). Our experiment showed that, as the thickness d' of the band-like film increases, $\left|\frac{dR}{dI}\right|$ decreases and approaches zero gradually. For the band-like film with thickness $d' \to d$, the phenomenon of $\frac{dR}{dI} < 0$ disappears gradually and the films will exhibit the quadratic *R-I* behavior (see Eq. 2). This result is in good agreement with the prediction by the RRN model (Ye et al., 1994b) and the phenomena observed in the other film systems (Yagil et al., 1992; Dubson et al., 1989). The increment of the thickness d' implies that more deposited Al atoms reach the glass surface before it is quenched and thus the average time for the atoms to reach the substrate and for the atom layers to be covered with the oil increases, which implies that the time period of the deposition atoms diffusing on the substrate increases and the real quenching effect reduces. Therefore, it is quite reasonable that the *R-I* behavior of the band-like film near the thick edge of the quasi-wedge film approaches that of the normal Al films deposited on glass surface.

CONCLUSIONS

From the preparation and dc conductivity measurement for a new aluminum thin film percolation system described above, we can draw the following conclusions:

1. A new Al film percolation system with a characteristic structure has been fabricated. The films were gradually quenched with the silicone oil gradually during the deposition and the characteristic microstructure of the film system is naturally formed.

2. The anomalous *R-I* behavior indicates that the films contain many defects and exhibit a characteristic percolation structure due to the immediate quenching process, which result in the anomalous hopping and tunneling effects in the system at very low currents.

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