Conductivity Measurements of a Metallic Diffuse-Fringe Film Percolation System

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We have fabricated a metallic diffuse-fringe film system by dc-magnetron sputtering method. The diffuse fringe structure of the film system is obviously observed when during the film deposition process the distance between the shutter slit and the substrate is large enough. An anomalous, non-linear $I-V$ behavior is found in the system and can be well explained by the Joule heating and hopping effects. The strong dependence between the conductivity behavior and the fringe indicates that the fringe effect is very severe in this inhomogeneous percolation system.

1. Introduction

Recently, many studies have been devoted to the investigation of rough surface structures and the rough surface effects of film systems. Several kinds of rough film systems have been fabricated [1 to 6]. It has been realized that the surface microstructure is essential to all the properties of thin films. Therefore, it is expected that the surface microstructure of a film can be controlled in more effective ways so that it has desirable properties.

In extra small physical systems (mesoscopic systems for instance), the fringe structure has a profound effect on all the electrical properties of the systems, and the microstructure must be characterized at nanometer or smaller length scales in order to gain an understanding of its physical origin [7 to 11]. In macroscopic systems, however, the fringe structure has rarely been studied so far. For example, the fringes of both flat and rough films are frequently considered to be ideal smooth edges and many effects of the irregular fringes are neglected [5, 6, 12 to 14].

In this paper, we report the preparation of a metallic diffuse-fringe film percolation system and its anomalous, nonlinear $I-V$ behavior. Our experimental result shows that the diffuse fringes of the film will obviously arise if the distance between the slit shutter and the glass substrate is large enough during the deposition process. The anomalous $I-V$ behavior is strongly dependent on the fringe structure and the rough surface of the film system, indicating the importance of the fringe effect of the system. A discussion on the physical origin of the characteristics is also presented.

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2. Experimental Details

The metallic diffuse-fringe film percolation system was deposited onto glass substrates by dc magnetron sputtering method. A slit of width $w$ ($\approx 0.1 \text{ mm}$) was carved in the shutter, which was fixed between the glass substrate and the metallic target. Other than ordinary, as a key step to prepare the diffuse-fringe films, we did not let the parameter $h$, the distance between the slit and the substrate, be zero. The thickness of the shutter is 0.05 mm, which is thin enough compared with the slit width $w$. The diameter of the target is 81.5 mm and the target–substrate distance equals 60 mm. Therefore, the micro-structure and the outline width $w_f$ of the film are very sensitive to both the parameters $w$ and $h$. During the deposition process, the sputtered atoms can only pass through the slit and then reach the substrate. The diffuse-fringe structure of the films is then formed since $h$ is not equal to zero. The diffuse-fringe films were deposited between four gold film electrodes by using this slit deposition (SD) method under an Ar gas pressure of 0.1 Pa at room temperature. The maximum deposition rate (since the rate is location dependent) was less than 0.1 nm/s. The length of each sample is 5.0 mm. The nominal thickness of the middle part of the films, obtained from the depositing rate, varies from 8 to $1.2 \times 10^2$ nm. The dc sheet resistance $R$ as a function of the dc current $I$ was measured in air with the four-probe method.

3. Results and Discussion

The scanning electron microscope pictures of two of our samples are shown in Fig. 1. It shows that, if we let the distance $h$ be very small during the deposition process, the fringe boundary of the film is very clear and the width of the film can be easily measured (see Fig. 1a), like the situation of conventional flat films. However, if $h$ is large enough, the diffuse fringes obviously appear and there is no distinct boundary in the diffuse-fringe film (see Fig. 1b). Therefore, only the outline width of the diffuse-fringe film, i.e., $w_f$, can be measured. We will show that, in the diffuse-fringe system, the fringe effect can no longer be neglected since the irregular and rough fringe has become an important part of the film and the $R$–$I$ behavior of the diffuse-fringe films is very different from that of the flat systems.

![Fig. 1. SEM photograph of the Au diffuse-fringe films. The width of the slit is $w = 0.20 \text{ mm}$; a) $h = 0.01$, b) 2.0 mm](image-url)
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306
305
304
303
302
301
300

I (mA)

0 4.0 8.0 12.0 16.0

Fig. 2. $R-I$ characteristic of one of the Au samples with $h = 2.0$ mm and $w = 0.20$ mm. The outline width of the sample is $w_f = 1.65$ mm. The solid line represents the fit $R = R_0 + B I^2$, $B = 1.80 \times 10^4$ V/A$^2$.

The dc current dependence of the resistance $R$ of the samples was systematically measured at room temperature. For the diffuse-fringe samples with $R_0 \leq 10^3$ Ω, where $R_0$ is the sheet resistance when the current approaches zero, as shown in Fig. 2 and 3, an anomalous nonlinear $R-I$ characteristic, which is significantly distinct from the various results of the flat film system, the bilateral rough film system, and the wedge-shaped film system [5, 6, 15] is observed: at low currents, the sheet resistance $R$ increases with the current $I$ (see Fig. 2), which can be well expressed as $R = R_0 + B I^2$, where $B$ is a constant. In Fig. 2, one finds $B = 1.80 \times 10^4$ V/A$^2$ for the Au sample. This quadratic nonlinear response, which is similar to that of the flat film systems [15], indicates that the local Joule heating effect is still the main process in the diffuse-fringe films at low currents.

When the current $I$ increases continuously and reaches the value $I_{m1}$, however, $R$ gets to its extreme value and then it begins to drop smoothly until $I$ equals $I_{m2}$ (see Fig. 3). If the current is further increased, then $R$ will increase again. We notice that this anomalous $R-I$ response is not a reversible behavior (see Fig. 4). The $R-I$ behavior changes greatly and is similar to that of the flat film systems [15] after the first measurement. This result suggests that the microstructure of the diffuse-fringe films is seriously damaged by the measurement.

For the samples with $R_0 > 10^3$ Ω, within our measurement range, an obvious peak-like $R-I$ behavior is observed and the point $I_{m2}$ does not appear, as shown in Fig. 5.

In order to find the relation between the distance $h$ and the $R-I$ behavior, we measured the $R-I$ behavior of three samples, as shown in Fig. 6. All the parameters and the deposition conditions of the three samples are identical except for the varying distance $h$. If $h$ is very small, the $R-I$ behavior is similar to that of the flat film systems (see Fig. 6a) [15]. This result is reasonable since the diffuse fringe structure will gradually disappear when $h$ approaches zero. However, if the distance $h$ is increased, the width of the film increases and then the film thickness decreases. Then the conductance...
of the film drops quickly. Also, the obvious diffuse fringe structure of the film is formed due to the increase of the distance $h$ and then an anomalous $R-I$ behavior appears (see Fig. 6b and c). The sensitive dependence described in Fig. 6 indicates that the $R-I$ behavior is really caused by the diffuse fringe structure of the films. From Fig. 6, one can see that the bigger the distance $h$ is, the smaller $I_{m1}$ and $I_{m2}$ will be. This result suggests that if $h \to 0$, $I_{m1}$ and $I_{m2}$ will approach infinity and the maximum and minimum of $R$ will disappear from the $R-I$ curve. This inference is in agreement with common ideas [15].
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Fig. 4. $R-I$ characteristic of an Au diffuse-fringe film with $h = 2.0$ mm, $w = 0.20$ mm, $w_f = 1.65$ mm. Dots: first measurement; squares: second measurement

Fig. 5. $R-I$ characteristics of an Ag diffuse-fringe film with higher resistivity; $h = 1.0$ mm, $w = 0.10$ mm, $w_f = 0.9$ mm
Fig. 6. $R-I$ characteristics of three Ag diffuse-fringe films. All the deposition conditions of the three samples are equal and $w = 0.10 \text{ mm}$; a) $h = 0.05 \text{ mm}$, $w_f = 0.10 \text{ mm}$; b) $h = 0.30 \text{ mm}$, $w_f = 0.58 \text{ mm}$; c) $h = 0.60 \text{ mm}$, $w_f = 0.80 \text{ mm}$

We propose that the $R-I$ characteristics described above are caused by the diffuse-fringe structure of the films since other systems do not exhibit such a behavior [5, 6, 15]. At lower current, the local Joule heating effect is the main contribution to the electrical process, which results in the quadratic $R-I$ behavior [15]. When the current increases
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beyond the current $I_{m1}$, however, a higher current passes through the weaker links in the diffuse fringe part of the film bringing about a large increase of the local temperature which is sufficient to excite local hopping. The hopping effect would reduce the current density of the links and, therefore, the sheet resistance. Therefore, a negative slope (i.e., $dR/dI < 0$) arises. When the current goes beyond $I_{m2}$, nearly all the hopping processes around the links become saturated and then the change of the sheet resistance is mainly caused by the Joule heating effect, which will result in a positive slope (i.e., $dR/dI > 0$) again. Since the nominal thickness of the fringes of the films (or the surface coverage fraction of the anisotropic percolation system) [5] changes linearly, continuously, and perpendicularly to the film length, the density distribution of the links changes gradually in this direction. Then both the location-dependent hopping (LDH) [6] and the Joule heating effect occur. Therefore, a smooth peak and valley appear in the $R$-$I$ curve (see Fig. 3, 4, and 6).

After the first measurement, the diffuse-fringe percolation structure of the fringes of the films is damaged (bond broken or bond linked). The bond-link effect, which may mainly happen among the bigger clusters, leads to the decrease of the film resistance $R$. Therefore, it is quite understandable that $R$ in the second experiment is less than that in the first experiment at low currents (see Fig. 4). Then the sheet resistance should be mainly contributed from the middle part of the films, which will simply result in the positive slope of the $R$-$I$ curve because of the Joule heating effect, just as in the flat film [15], and $I_{m1}$ and $I_{m2}$ will no longer appear.

Both theoretical and experimental researches prefer a perfect and smooth film fringe rather than an “irregular” or “rough” one, since on the theoretical side [12, 13, 16], the irregular fringe is very difficult to be managed and, on the practical side, the fringe effect is actually small in flat and rough film systems [5, 13, 15]. However, in the diffuse-fringe film system, the “fringe” and the “main part” of the film can no longer be divided from each other and there is no obvious boundary between them. Therefore, the fringe structure is no longer a “defect” or a “nonideal” structure, but is an important part of the film. The various properties of the diffuse-fringe system should be seriously influenced by the fringe effect, which can be both advantageous and disadvantageous (Fig. 3, 4, and 6). We find that the critical behavior of this new type of percolation system is also related to the diffuse fringe structure. A detailed description of the phenomenon will be published separately.

4. Conclusions

In summary, we have measured the dc $R$-$I$ behavior on a diffuse-fringe film percolation system deposited on glass substrates by the SD method. The SEM photograph of the samples show that if the parameter $h$, the distance between the slit and the substrate is large enough, an obvious diffuse fringe structure appears. An anomalous nonlinear $R$-$I$ behavior, which is sensitive to the parameter $h$ and the sheet resistance $R_0$, is found. This anomalous nonlinear response is due to both the location dependent hopping and Joule heating effects happening in this anisotropic diffuse-fringe percolation system, since the conventional flat film and rough film systems do not exhibit such a property. We believe that the above result is very helpful for preparing and studying other new rough film systems (the diffuse-fringe multilayered rough film system, for instance). This subject remains an interesting topic for further study by both theoretical and experimental methods.
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References