Conductivity and dielectric constant in a wedge-shaped Pt-film percolation system

Gao-xiang Ye 1, Yu-qing Xu, Hong-liang Ge, Zheng-kuan Jiao, Qi-rui Zhang
Department of Physics, Zhejiang University, Hangzhou 310027, China

Received 8 June 1994; revised manuscript received 3 August 1994; accepted for publication 7 December 1994
Communicated by J. Flouquet

Abstract

A wedge-shaped Pt-film percolation system was prepared by the magnetron sputtering method. A new nonlinear $I-V$ behavior was found and can be well explained by the location-dependent hopping effect. Power-law behavior, $\sigma(\omega) \propto \omega^x$ and $\varepsilon(\omega) \propto \omega^{-y}$, is observed near the percolation threshold. The exponents $x$ and $y$ are found to be $0.87 \pm 0.04$ and $0.08 \pm 0.03$, respectively, in agreement with the general scaling relation $x + y = 1$.

All properties of thin films, particularly ultrathin films, may be affected sensitively by the surface microstructure below submicron length scales. Many interesting and peculiar phenomena have been observed in rough thin film systems [1–4]. It has been realized that it is not necessary to prepare films with very smooth surfaces since the rough surface effect can be both advantageous and disadvantageous. In fact, the surface structure of films is also essential for multilayered films, quantum wells and superlattices although the physical origins (the giant magnetoresistance effect for example) [5] have not been well studied so far. Therefore, considerable effort has recently been expended on investigating the conditions for creating a particular morphology, detecting universality relationships, and studying electrical properties.

One-side rough films, in which only the up surfaces of the films are roughened, are frequently fabricated by the ion bombardment method and the nonequilibrium growth technique and scaling behavior has been observed in these systems [6,7]. Recently, two-side rough thin film systems (i.e., both the up and down surfaces of the films are uneven) have been produced by using disordered and stepped substrates and many interesting characteristics have been found in these systems [3,4]. However, to control the surface morphology more effectively so that the film has desirable electrical, optical, magnetic, or mechanical properties, much effort is still needed.

In this paper, we report the preparation of a wedge-shaped Pt-film percolation system and its peak-like nonlinear $I-V$ behavior. Our experimental result shows that this nonlinear $I-V$ characteristic is strongly dependent on the wedge angle and the crystallinity of the film. We will also show that, although the wedge-shaped film system does not belong to both the flat lattice-percolation system and the flat continuum-percolation system [8,9], its critical behavior can be well described by power laws and the general scaling relation still holds in this anisotropic system. A discussion of the physical origins of the characteristics is also presented.

1 Permanent address: Department of Physics, Hangzhou University, Hangzhou 310028, China.
The wedge-shaped Pt-film percolation system was deposited onto glass substrates by dc- or rf-magnetron sputtering methods. During the deposition process, each substrate was placed in such a way that the distances from the target shutter to the different regions of the substrate were obviously different. Therefore, a gradient deposition rate was formed. The wedge films were deposited among four gold film electrodes by using this location-dependent deposition rate (LDDR) 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 0.2 nm/s. The size of each sample was 2.0 mm × 22.0 mm. A schematic drawing of the samples is shown in Fig. 1. d1 and d2 represent the thicknesses of the thicker and thinner ends of the wedge-shaped Pt film, respectively. Within the film resistance range of 3 kΩ to 1 MΩ, the nominal thickness d2 of the samples varies from 6 nm to 11 nm. Therefore, the average percolative channel width (i.e., the hopping length) at the thinner ends of the samples should be of the order of d2 [9,10]. Here, we propose that the wedge-shaped film be characterized by the parameter \( \theta = \frac{d_1}{d_2} \) and the film resistance \( R \).

The dc sheet resistance \( R \) as a function of the dc current \( I \) was measured in air with the four-probe method. The ac conductance and ac capacitance were measured simultaneously in air and at room temperature using a Yokogawa Hewlett-Packard 4275A multifrequency LCR meter with four-probe geometry. A standard resistor and a standard capacitor were used to ensure that the observed result was correct. The frequency range covered by this experiment was 10 kHz to 10 MHz. The peak-to-peak voltage was 0.4 V and no heating effect was observed.

The dc \( I-V \) characteristic was measured and three distinct regimes of \( R_0 \) were found, where \( R_0 \) is the sheet resistance when the current \( I \) approaches zero.

For the wedge-shaped samples with lower resistivity, the \( R-I \) behavior is shown in Fig. 2. The \( R-I \) relation can be well expressed as

\[
R = R_0 + BI^2,
\]

where \( B \) is a constant. In Fig. 2, one finds \( B = 8.486 \times 10^5 \) V/A^3 for the sample with \( \theta = 2.6 \). This behavior is similar to that of the flat film systems [10]. It should be noted that since these films are relatively thicker, the wedge structure effect is very weak and the \( I-V \) behavior could not be obviously affected. Thus, the above result is reasonable.

For the samples with higher sheet resistance and again \( \theta = 2.6 \), as shown in Figs. 3 and 4a, an obvious peak-like \( R-I \) behavior is observed: at low currents, the sheet resistance \( R \) increases with the current \( I \). When \( I \) equals \( I_m \), however, \( R \) reaches its maximum value and it then begins to drop smoothly. We notice that, although the sheet resistance \( R \) and current \( I_m \) would be slightly changed after the first measurement, the peak-like \( R-I \) relation will not be altered apparently. From Fig. 3, one finds that the changes of \( R \) and \( I_m \) are relatively small (\( \Delta R/R < 1\% \); \( \Delta I_m/I_m < 5\% \)), which suggests that the wedge-shaped structure effect would not be affected greatly by the first lower current measurement. However, if more measurements are done, the film resistance (for fixed \( I \)) will further increase and seems to go towards a saturation, then the peak will gradually disappear from the \( R-I \) curve, indicating that, at that time, the location-dependent percolation structure has been seriously damaged by the Joule heating.

---

**Fig. 2.** \( R-I \) characteristic of one of the thicker Pt wedge-shaped films with \( \theta = 2.6 \). The solid line represents the fit \( R = R_0 + BI^2 \), \( B = 8.486 \times 10^5 \) V/A^3.
just like for the annealing process (see below). Figs. 3 and 4 also show that the larger $R_0$, the smaller $I_m$ will be. Therefore, for the sample with small $R_0$, the peak cannot be observed since $I_m$ is too big and the film will be seriously damaged before the peak appears (see Fig. 2).

On the other hand, when the wedge angle increases (see Fig. 5a, $\theta = 5.0$), the peak-like $I-V$ relation still holds, but the current $I_m$ becomes much smaller. This result suggests that if $\theta \to 1$, i.e., the film becomes a flat one, $I_m$ will approach infinity and the peak will disappear from the $R-I$ curve. This inference is in agreement with the common ideas [10].

It is noticed that the peak-like $R-I$ behavior is strongly dependent on the crystallinity and the percolative structure of the film. After annealing in a vacuum of better than $4 \times 10^{-3}$ Pa at $600^\circ$C for 1.5 hours, the samples exhibit a $R-I$ behavior $dR/dI > 0$ and the peaks have disappeared from the $R-I$ curves (see Figs. 4b and 5b), indicating that the hopping conductivity is important in the $R-I$ behavior described in Figs. 3–5.
For films with $R_0 > 50 \text{k}\Omega$ ($\theta = 2.6$), within our measurement range, $R$ will simply decrease with $I$, i.e., $dR/dI < 0$, and the reversible characteristic becomes poorer, just as for the flat film [10]. This result indicates that the hopping effect becomes important in the electrical process.

We propose that the $R-I$ characteristics described above are mainly caused by the wedge-shaped structure of the films since the flat systems do not exhibit such behavior [10]. At lower current, the local Joule heating effect is the main contribution to the electrical process, which results in the phenomenon of $dR/dI > 0$ [10]. When the current increases, however, that a higher current passes through the weak links in the thinner part of the film would bring 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 then the sheet resistance. Therefore, a negative slope (i.e., $dR/dI < 0$) arises. Since the thickness of the film changes linearly and continuously along the length, the links are distributed gradually and a location-dependent hopping (LDH) effect occurs. Then, a very smooth peak appears in the $R-I$ curve. After the annealing process, the percolative structure as well as the crystallinity of the samples are changed and the islands in the films gradually merge. Then the number of the links or junctions is greatly reduced. Therefore, the hopping effect in the film becomes weak and $dR/dI < 0$ will no longer occur.

In the case of flat high-resistivity metallic films deposited on glass substrates, it is shown that weak links are usually short circuited by a hopping process initiated by the temperature rise at the hot spots [10]. The resistivities of the thinner ends of our samples are also in this resistivity interval and then the hopping process should occur in the wedge-shaped system. In particular, we expect to see what the wedge-shaped structure effect on the hopping process is. For this purpose, it is meaningful, e.g., to measure the temperature dependence of the resistivity, to distinguish between the anisotropic effect and the surface effect, and to find the relationship between the parameter $\theta$ and the cluster size distribution. This work is presently in progress and a detailed description will be published separately.

The ac conductance $\sigma(\omega)$ and capacitance $C(\omega)$ (which is proportional to the real part of the dielectric constant $\epsilon(\omega)$) measurements for the wedge-shaped films were done at room temperature and the results are plotted on a logarithmic scale as a function of the frequency $f = \omega/2\pi$ in Figs. 6 and 7.

Fig. 6 shows a flat frequency response for samples far from the percolation threshold $p_c$. However, when approaching $p_c$, a power-law behavior,

$$\sigma(\omega) \propto \omega^x, \quad \epsilon(\omega) \propto \omega^{-\gamma},$$  \hspace{1cm} (2)

is observed in a certain frequency range, which is consistent with the results of the isotropic percolation sys-
From the experimental data of the ac conductance and the ac capacitance in Figs. 6 and 7, the critical exponents $x$ and $y$ are determined to be $0.87 \pm 0.04$ and $0.08 \pm 0.03$, respectively. Then, $x + y = 0.95 \pm 0.07$, which satisfies the general scaling relation [12]

$$x + y = 1.$$  \hspace{1cm} (3)

The exponents $x$ and $y$ obtained above are almost the same as the results of flat film percolation systems [8,9,11]. This fact shows that the anisotropic structure of the wedge-shaped film does not affect the critical behavior of the ac conductivity and the dielectric constant and the exponents $x$ and $y$ are universal. We notice that Eqs. (2) and (3) were derived by Bergman and Imry without making any scaling assumptions [12]. To explain the power-law behavior, several factors should be taken into account: (1) the intercluster polarization effect, resulting in the electron-electron interactions and the capacitive coupling between the percolation clusters, (2) the anomalous diffusion model describing the time delay and the consequences of the fractal nature of each individual cluster, (3) the significant `current leakage' near the corners of clusters due to the fringe effect [11]. Unfortunately, at present, a theory containing all of these factors does not exist. We believe that, near the percolation threshold, the factors described above are still the main electrical processes in the wedge-shaped film since the percolative structure is still available in the thinner part of the film. Therefore, the power-law behavior and the general scaling relation should be satisfied in this anisotropic system. However, to reach a final conclusion, more experimental data for the ac conductivity and dielectric constant for the anisotropic percolation system are needed.

In summary, we have measured the dc resistance $R$, ac conductivity $\sigma(\omega)$, and ac dielectric constant $e(\omega)$ on a wedgeshaped Pt-film percolation system deposited on glass substrates by the LDDR method. A peak-like nonlinear $R-I$ behavior, which is strongly dependent on the wedge angle and the crystallinity of the film, is observed and can be well explained by the LDH effect. Our experimental result shows that the power-law behavior, $\sigma(\omega) \propto \omega^x$ and $e(\omega) \propto \omega^{-y}$, still holds in the system. The exponents $x$ and $y$ are found to be $0.87 \pm 0.04$ and $0.08 \pm 0.03$, respectively, in agreement with the general scaling relation $x + y = 1$, which supports the prediction that the two exponents might actually be universal.

We have benefited greatly from helpful discussions with Professor X.J. Zhang, Dr. M.C. Tan, and X.M. Tao. The experimental contribution of Engineer H.L. Wang is gratefully acknowledged. This work was supported by the Zhejiang Provincial Natural Science Foundation of China (Grant No. 193054).

References