Evidence of anomalous hopping and tunneling effects on the conductivity of a fractal Pt-film system

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The temperature dependence of the conductance and the nonlinear electrical response of a Pt-film percolation system, deposited on fracture surfaces of α-Al_2O_3 ceramics, have been measured over three decades of sheet resistance. We find that in the temperature interval of $T=77-300$ K, the resistance temperature coefficient $\beta=(1/R)dR/dT$ is not a constant, which is different from that for flat films. A dc I-V characteristic which strongly depends on the thickness of the film is found and it can be interpreted as a competition among the local Joule heating, hopping, and tunneling effects. The third-harmonic measurement suggests that the critical exponent comes from $1/f$ noise, which obeys the power-law dependences $S_R \propto (p-p_c)^{-\kappa}$, $R \propto (p-p_c)^{-t}$, and then $S_R \propto R^w$ with $w=\kappa/t$, where S_R is the mean square of resistance fluctuations, p the surface coverage fraction, and p_c its percolation critical value. We find that $w=0.45\pm0.06$, which is lower than the flat-film exponent. This result indicates that the tunneling and hopping effects in the fractal samples are much stronger than that of flat films.

I. INTRODUCTION

There is a very sensitive and complex dependence of thin-film microstructure on growth conditions. The microstructure at or below submicron length scales frequently has a profound effect on all physical properties (magnetic, electrical, mechanical, optical, etc.) of the thin-film system.[1–4] In fact, the self-affine fractal structures of films are also essential to various multilayered films, quantum wells, and superlattices, although its physical origins are less studied so far.

Usually, roughness thin films are made by the ion-bombardment method and the nonequilibrium growth technique. These films have been proven to have the self-affine fractal surfaces and can be well described by scaling theory.[1,5,6] On the other hand, a type of roughness thin films, in which both the up and down surfaces are uneven, can be made by using the roughness substrates, and many interesting phenomena have been found.[4,7–9] These films, particularly ultrathin films, also have the self-affine fractal structures. The experimental results indicate that, just as the flat substrates, the self-affine fractal substrates may also be very useful for both fundamental and practical purposes.[4]

In this paper, we report our measurements of the temperature dependence of resistivity and the nonlinear dc I-V behavior of the Pt-film percolation system deposited on fracture surfaces of α-Al_2O_3 ceramics. We find that, over three decades of sheet resistance, the films have a positive temperature coefficient of resistance in the interval $T=77-300$ K. The nonlinear electrical response is much different from that of the flat-metal films. The third-harmonic generator B is found to scale as R^{2+w} with critical exponent $w=0.45\pm0.06$, which indicates that the hopping and tunneling effects in this system are much stronger than those of the flat films.

II. SUMMARY OF THE THEORY AND ITS APPLICATION TO DC MEASUREMENT

The microgeometry of a percolative film can be probed by the $1/f$ noise measurement, in which the fourth moment of the current distribution is measured.[10–14] The film system can be considered as a random resistor network with total sheet resistance R, carrying a current I, made up of elements of resistance r_α carrying currents i_α. By the conservation of energy, we have

$$I^2R=\sum r_\alpha i_\alpha^2 \ . \tag{1}$$

Using Tellegen's theorem,[13] the relative resistance noise S_R is given by

$$S_R=(\langle \delta R^2\rangle)/R^2=\left[\sum i_\alpha^4\langle \delta r_\alpha^2\rangle\right]\Big/\left[\sum i_\alpha^2 r_\alpha\right]^2 , \tag{2}$$

which probes the fourth-current distribution moment. Rammal *et al.*[13] have introduced a nonuniversal exponent κ that describes the divergence of the noise power

$$S_R \propto (p-P_c)^{-\kappa} , \tag{3}$$

where p is the surface coverage fraction and p_c is its percolation critical value. The film resistance obeys another power law

$$R \propto (p-p_c)^{-t} . \tag{4}$$

Combining Eq. (3) with Eq. (4) yields

$$S_R \propto R^w , \tag{5}$$

where $w=\kappa/t$.

Consider a resistor network carrying an ac current $I=I_0\cos(\omega t)$ with each element α carrying a current $i_\alpha\cos(\omega t)$. Assuming the resistors have a positive temperature coefficient of resistance and the amplitude of the local temperature oscillation is proportional to the amplitude of the Joule power generated, then Joule heating would cause the sample's resistance to oscillate at 2ω:[12]

$$R = R_0 + \Delta R \cos(2\omega t + \phi) , \quad (6)$$

the amplitude of the resistance is proportional to the fourth moment of the current distribution

$$\Delta R = \beta h(\omega)/(I^2) \sum i_\alpha^4 r_\alpha^2 , \quad (7)$$

where $\beta = (1/R)(dR/dT)$, $h(\omega)$ is a function of frequency but not of α, and ϕ is the phase shift between the heat production and the local temperature. The voltage across the sample is given by

$$V = IR \approx I_0 R_0 \cos(\omega t) + (\tfrac{1}{2}) I_0 \Delta R \cos(3\omega t + \phi) . \quad (8)$$

The third-harmonic coefficient is

$$V_{3f} \propto I_0 \Delta R \propto (1/I_0) \sum i_\alpha^4 r_\alpha^2 , \quad (9)$$

which is related to the fourth-current distribution moment. Hence, using Eq. (2), the quantity S_R can be written

$$S_R \propto V_{3f}/(I_0{}^3 R^2) , \quad (10)$$

where, as usual, we take $r_\alpha = r$ for all α.[12] Therefore, V_{3f} should scale as

$$B \equiv V_{3f}/I_0^3 \propto R^{2+w} . \quad (11)$$

Now, let ω approach zero, then Eq. (8) becomes

$$R = R_0 + BI^2 , \quad (12)$$

where R_0 is the sheet resistance when I approaches zero. Then, the coefficient B can be obtained by the dc measurement. It has been proved that, if $\omega \to 0$, the value of B obtained by the ac measurement will approach the dc result and, furthermore, the critical exponent w is independent on frequency ω although the coefficient B depends on the frequency.[12,14] Therefore, using Eq. (12) to measure the critical exponent w is an ideal method for its simplicity and accuracy. The third-harmonic-generation method described above can be used for measuring samples with lower sheet resistance.[14,15]

III. EXPERIMENT

The sample preparation method has been described in our previous work.[4,7,8] The fractal dimension of the substrates was $D_0 = 2.20 \pm 0.06$. The films were deposited by the dc-magnetron-sputtering method among four gold-film electrodes. The size of each sample was 6.0×2.0 mm^2. The dc sheet resistance R as a function of the temperature T and the dc current I was measured with the four-probe method.

IV. RESULTS AND DISCUSSION

A. The temperature dependence of the resistance

Figure 1 contains the conductance σ vs $1/T$ curves for five Pt fractal films from 77 to 300 K. In a wide span of resistances (about 200–200 000 Ω), a good linearity of the log conductance vs $1/T$ plot is found, and the films have a positive temperature coefficient of resistance, indicating that, at low current ($I = 10\ \mu$A), the conductivities are metallic. Such behavior is similar to that of flat-metal films although the substrates are totally different.[16] However, the coefficient β is not a constant, which is contrary to that of the flat system.[14] The results of twelve samples have been fitted by $\sigma = \sigma_0 \exp(-T/T_0)$. Where $T_0 = \varphi/k$, k is the Boltzmann's constant, and φ is the activation energy. The optimal values for T_0 are listed in Table I. It is found that the samples could be divided into two groups. The films belonging to the lower resistance group have larger values of T_0 and those belonging to the higher group have smaller values of T_0. The activation energies of the higher resistance samples show no trend with the sheet resistance R, providing evidence for

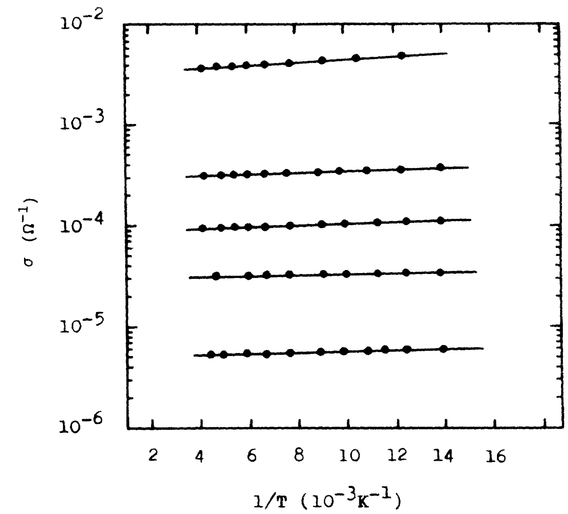

FIG. 1. Conductance vs reciprocal temperature for platinum fractal films of different thicknesses.

TABLE I. Parameter T_0 obtained by fitting the experimental temperature dependence of the dc conductance $\sigma(T) = \sigma_0 \exp(-T_0/T)$.

Sample	$R(\Omega)$	T_0 (K)
1	272	37
2	350	35
3	2 168	12
4	2 684	14
5	3 198	16
6	3 350	18
7	4 753	13
8	4 926	13
9	6 072	11
10	10 595	14
11	31 110	10
12	188 000	13

the importance of the hopping and tunneling conductivities in this resistance span (see Table I). This is contrary to the results of flat-metal films.[16,17] We propose that the sharp drop (compared with the flat-metal films) of T_0 is because of the fractal structures in the films. For the lower resistance samples, i.e., $R < 1$ kΩ, the films are relatively thicker and the fractal structures on the substrates are then submerged. However, when the film resistance is beyond 1 kΩ,[7] the self-affine fractal microstructure will begin to appear on the films and, will apparently change the dependence of conductance on temperature.

For ultrathin fractal films, i.e., $R > 1$ MΩ, the resistances showed large fluctuations in time (the current used is 10 μA). The amplitudes of the fluctuations are seemingly not strongly dependent on the temperature. A detailed description of the phenomenon that occurs during the approach to zero conductance will be published separately.

B. *I-V* behavior measurements

The *I-V* characteristic was measured and three distinct regimes of the sheet resistance R_0 were found. For the thick film, i.e., $R_0 < 10$ kΩ, the *R-I* behavior is similar to that of the flat film, although their substrates are quite different.[14,15] As shown in Fig. 2, at low currents the film shows Ohmic behavior. At higher currents, however, the resistance increases with the current, i.e., $dR/dI > 0$. This reversible *R-I* relation can be well fitted by Eq. (12). If the current is further increased and beyond the breakdown current I_c,[15] then an irreversible and discontinuous change will occur (in Fig. 2, $I_c \approx 10$ mA). The nonlinear response described above has been well studied and is generally interpreted in terms of a heating and melting process of the hot spots (or links) due to the local Joule heating.[12]

It is very reasonable that the thick fractal film has the quadratic *R-I* behavior, just as the flat film does. In fact, when the film thickness increases, the fractal structure of the substrate will be covered and then the microstructure in the film will disappear gradually. Finally, the sample becomes a thicker and smoother film. Thus, all the characteristics of the thick fractal films should be similar to those of the flat films. Our previous work has verified this expectation.[4,7,8]

It has been proven that the power-law behaviors, existing in the lattice-percolation and continuum-percolation systems, would not be affected apparently by the fractal geometries of the substrates.[4] Fitting the experimental data in Fig. 2 by the Eq. (12), one finds $B = 2.0\times10^6$ (V/A^3). Using this method, the coefficient B of different samples with different sheet resistances are obtained and the scaling of B as a function of the sheet resistance R_0 is shown in Fig. 3. Again, a power law with the critical exponent $w = 0.45\pm0.06$ is well defined up to $R_0 = 10$ kΩ. This value of w is much smaller than the flat-film exponent[10,14] and the theoretical predictions in two-dimensional system.[13,18–20]

The *R-I* behavior of samples with resistances higher than 10 kΩ cannot be fitted by Eq. (12), as shown in Fig. 4. At low current, the sheet resistance drops with the increase of I; when the current further increases and reaches a critical value, however, the resistance starts to increase. From Fig. 4, one can see that R (as well as R_0) will be increased after the first *R-I* measurement. Therefore, this *R-I* relation corresponds to an irreversible behavior. The relative change of the resistance $t = \Delta R/R$ in Fig. 4 is about 8%.

For films with resistances higher than 30 kΩ (see Fig. 5), within our measurement range, the resistance will simply decrease with the current, i.e., $dR/dI < 0$ and, for a fixed current, the value of dR/dI (absolute value) would increase when R_0 becomes larger. This conclusion is also true for the flat metallic films.[15] However, in Fig. 5, dR/dI will approach zero and seems to be unchanged with further increasing of the current, which is contrary to the situation of the flat films.[15] Again, the *R-I* behavior described in Fig. 5 cannot be well repeated and

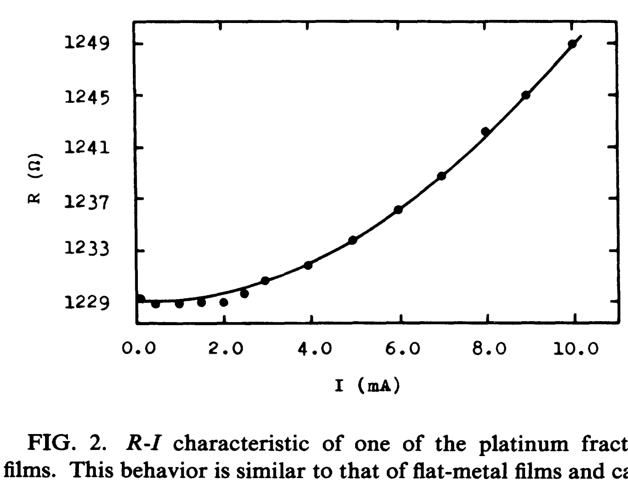

FIG. 2. *R-I* characteristic of one of the platinum fractal films. This behavior is similar to that of flat-metal films and can be interpreted in terms of the Joule-heating effect. Dots are the experimental data and the solid line represents the fit $R = R_0 + BI^2$, $B = 2.0\times10^6$ (V/A^3).

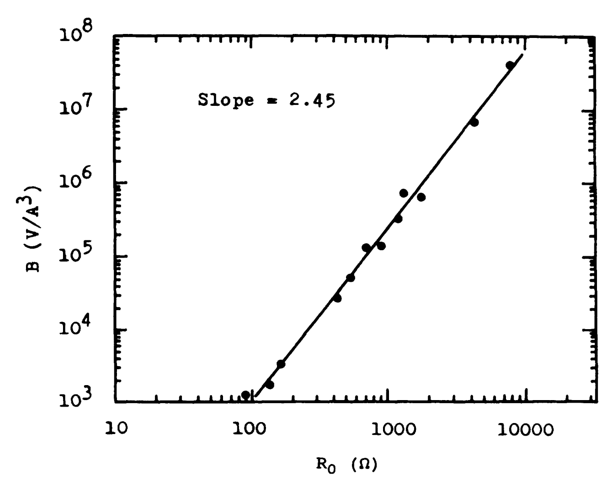

FIG. 3. The scaling of B for platinum fractal films, $2+w = 2.45\pm0.06$.

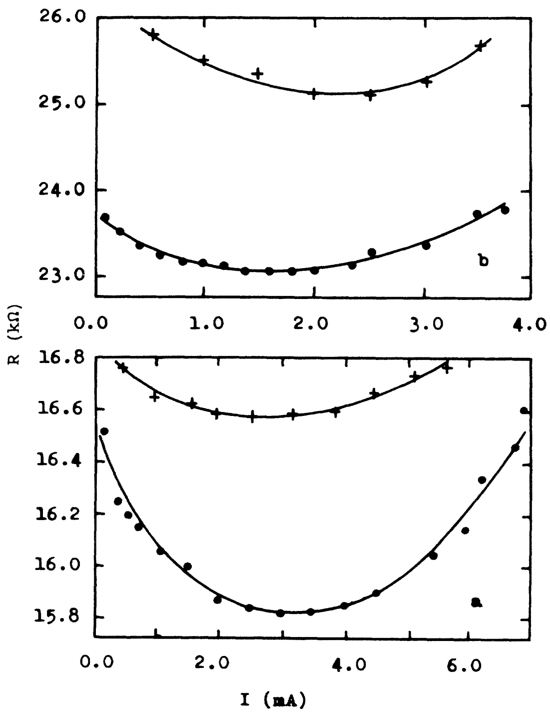

FIG. 4. R-I characteristics of platinum fractal films with $R_0 > 10$ kΩ. Dots and plus signs represent the first and the second R-I measurement results, respectively; the solid lines are a guide to the eye.

the sheet resistance R will increase greatly after the first R-I measurement (see Fig. 6). The relative change of the resistance is much larger than that of the low resistance samples (in Fig. 6, t is around 15%), which means that, for a certain current, the microstructure of the high resistance films changes much more than that of the low resistance samples.

That the nonlinear I-V characteristics in Figs. 4 and 5 correspond to irreversible behavior indicates that the hot-spot melting process due to Joule heating occurs in these cases. Therefore, the coefficient B cannot be obtained by the third-harmonic method since, in these cases, the conductivity of the samples is not purely metallic due to the stronger hopping, tunneling, and the irreversible melting process, and the third-harmonic signal is weaker. In fact, such a phenomenon also exists in the samples with resistance $R_0 < 10$ kΩ although the influence on that R-I characteristic is relatively weaker and can be omitted (see Fig. 2). To the best of our knowledge, the electrical breakdown behavior of thin-metal films with resistance $R_0 > 10$ kΩ has not been well studied for both smooth and roughness films. It is obvious that the phenomenon described in Figs. 4 and 5 cannot be simply explained by the Joule-heating effect[14] and the two-dimensional theoretical models.[13,18–20]

We proprose that the lower critical exponent w and the anomalous R-I behavior is caused by the rough surface of the substrate. Since the substrate has the self-affine structure, a huge number of defects and fractal structures would exist in the film. Therefore, many weak links, just as various metal-insulator-metal (MIM) tunneling junctions, are constructed in the films. When a high current passes through a weak link, the local temperature change is sufficient to excite local hopping and breakdown of the MIM tunneling junction. Both the hopping and tunneling effects would reduce the current density of the link and the sheet resistance. The higher the current is, the more the amount of the resistance will be reduced, which causes a weaker third-harmonic component [see Eq. (12)], and hence the critical exponent w becomes smaller. According to this analysis, the measured value of w, which is smaller than all the results of flat-film systems,[14] indicates that the hopping and tunneling effects in fractal films are much stronger than those of the flat films. This conclusion is very reasonable since the weak links (or tunneling junctions) in the self-affine films are much more plentiful than that in the flat systems and the film microgeometries are different from each other. We also propose that the phenomenon of $dR/dI < 0$ (see Figs. 4 and 5) is mainly caused by the short circuit due to the tunnel-

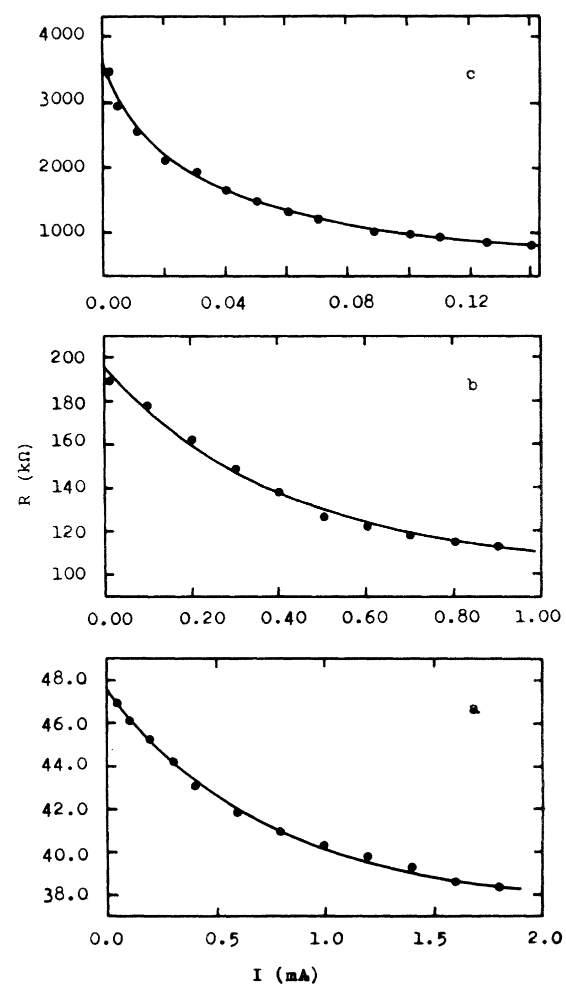

FIG. 5. R-I characteristics of ultrathin platinum fractal films due to hopping and tunneling effects. Dots represent the experimental results and the solid lines are a guide to the eye.

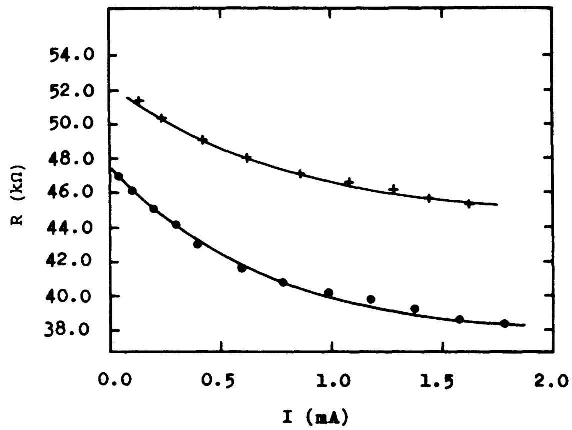

FIG. 6. R-I characteristics of the Pt films with $R_0 > 30$ kΩ. Dots and plus signs represent the first and the second R-I measurement results, respectively; the solid lines are a guide to the eye.

ing and melting processes at the hot spots. This effect will become more serious in measuring the samples with higher sheet resistance. Therefore, the higher the sheet resistance, the bigger the value of dR/dI will be (within our measurement span). When all the weaker MIM junctions have been broken down, then the value of dR/dI will decrease, as shown in Figs. 4 and 5. This result is quite different from that of the flat films, in which the value of dR/dI will increase with the current.[15]

V. CONCLUSION

In summary, we have measured systematically the dependence of conductance on temperature and the I-V behavior on a Pt-film percolation system deposited on the fractal surfaces of α-Al_2O_3 ceramics. The measurements provide a wide span of resistance and current: three decades of both R_0 and I. Our measurement shows a good linearity of the log conductance vs $1/T$ plot at $T = 77-300$ K and the films have a property of metallic conductivity. However, the resistance-temperature coefficient β is not a constant ($\beta \propto T^{-2}$), which indicates that, in this film-thickness interval, the temperature dependence of the conductance could be affected distinctly by the self-affine structure of the substrate. From the R-I behaviors, three distinct regimes of sheet resistance are found (Figs. 2, 4, and 5). In different regimes, the I-V relation has different behaviors. By using a new dc technique, a power law with critical exponent $w = 0.45 \pm 0.06$ is well defined up to $R_0 = 10$ kΩ; beyond this value, the tunneling and hopping processes would become so serious that their influence on the R-I relation cannot be omitted. Therefore, the current interval, in which dR/dI is a negative value, appears obviously. The above results can be interpreted under the assumption that all the Joule heating, hopping, and tunneling effects would make contributions to the R-I characteristics of the films simultaneously. However, a flexible theoretical model, including all the aspects of Joule heating, hopping, and tunneling, is still lacking.

The results above show us that the fractal substrates have various influences on the electrical properties of the metal films. We have found that these influences are strongly dependent on the film thickness. Further research on the physical origins of these effects is still needed.

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