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Third-harmonic coefficient in a Au-film percolation system deposited on fracture surfaces of α-Al_2O_3 ceramics

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The third-harmonic coefficient B_0 in a Au-film percolation system deposited on α-Al_2O_3 fracture surfaces by dc magnetron sputtering method is studied. The critical current I_c, coefficient B_0, and the relation between I_c and B_0 are measured as functions of the zero-power sheet resistance R_0. The power-law behaviors $I_c \propto R_0^{-\alpha}$ and $I_c \propto B_0^{-x}$ are observed in the system and the exponents α and x are found to be 0.51 ± 0.05 and 0.24 ± 0.08, respectively, which are different from those of other percolation systems and predictions of present theories applied to two-dimensional systems. The physical interpretation of the results and the universality discussion are also presented.

Recently, much experimental and theoretical work about the rough-surface effect in thin films has been carried out. Several kinds of rough thin-film systems, including the continuum-percolation film, lattice-percolation film, wedge-shaped film, one-side rough film, and bilateral rough film, have been fabricated in experiment and many anomalous phenomena, such as electrical breakdown behavior, weak localization, $1/f$ noise, etc., are observed.[1-8] These physical phenomena are believed to be strongly related to the rough-surface effect, which becomes very important if the amplitude of the rough-surface undulation approaches or goes beyond the order of the film thickness.

In thin-film percolation systems, the critical current I_c is found to vanish above the percolation threshold p_c as $I_c \propto (p-p_c)^y$, where p is the surface coverage fraction of the conductor. The zero-power sheet resistance R_0, which equals the value of the sheet resistance R when the current I approaches zero, is described by another power law, $R_0 \propto (p-p_c)^{-t}$. Combining these two power laws yields

$$I_c \propto R_0^{-\alpha} , \tag{1}$$

where $\alpha \equiv y/t$ is a critical exponent, which is extremely sensitive to the detail of the film microgeometry.[3] The theoretical work on predicting this exponent is far from satisfactory so far.[3,9-11] Since the divergence of the mean square of resistance fluctuation S_R obeys $S_R \propto (p-p_c)^{-\kappa}$, therefore,

$$S_R \propto R_0^{w} , \tag{2}$$

where $w \equiv \kappa/t$. The theoretical analysis of the random resistor network model gives[12]

$$B \equiv V_{3f}/I_0^3 \propto R_0^{2+w} , \tag{3}$$

where V_{3f} is the third-harmonic amplitude, I_0 is the amplitude of the ac current, and B is the normalized third-harmonic coefficient. Then, the exponent w can be obtained by measuring the values of B and R_0. Yagil, Deutscher, and Bergman[3] have introduced a critical exponent x that describes the power law

$$I_c \propto B^{-x} \tag{4}$$

and predicted that x is almost insensitive to fine details of the microgeometry of the films. On the other hand, 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

$$R = R_0 + B_0 I^2 . \tag{5}$$

Recently,[8,12] it is proposed that B_0 and R_0 also satisfy

$$B_0 \propto R_0^{2+w} , \tag{6}$$

and the exponent w obtained from Eq. (6) should be equal to the ac result. Therefore, w can also be obtained by a simple method, i.e., measuring the coefficients B_0 and R_0.

In this paper, the coefficient B_0 in a Au-film percolation system deposited on α-Al_2O_3 fracture surfaces by dc magnetron sputtering method is studied. We find that the power laws $I_c \propto R_0^{-\alpha}$ and $I_c \propto B_0^{-x}$ are still held in the Au rough-film system and the values of α and x, however, are different from those of the other film systems. This result indicates that both α and x would be strongly affected by the rough-surface effect (or the surface microgeometry).

In two- or three-dimensional percolation systems, the physical properties are usually characterized by universal power laws [$R_0 \propto (p-p_c)^{-t}$, for instance], where the critical exponents depend only on the dimensionality. Some properties [Eq. (2) for example], however, show highly nonuniversal behavior, where the exponents are extremely sensitive to fine details of the microgeometry.[13] We will emphasize that, since the dimensionality (i.e., the fractal dimension) of the bilateral rough-film system is strongly related to the microgeometry, the present universality wording is inadequate in describing the bilateral rough system.

The sample preparation method has been described in our previous work.[14] The fractal dimension of the α-Al_2O_3 substrates was $D_0 = 2.20 \pm 0.06$. The Au-film percolation samples, i.e., the bilateral rough films, were de-

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posited by the dc magnetron sputtering method among four gold-film electrodes with room temperature. The sputtering target employed was a metallic Au disk (purity 99.96%). The films were deposited under an Ar gas pressure of 0.1 Pa and at a rate of 0.1 nm/sec. The size of each sample was 6.0×2.0 mm^2. The dc sheet resistance R as a function of the dc current I was measured with the four-probe method.

The R-I characteristic of a Au rough sample is shown in Fig. 1. At low currents, the Ohmic behavior (i.e., R is a constant) is observed. At higher currents, however, the sheet resistance increases with the current, i.e., $dR/dI>0$. In this current regime, the R-I relation can be well fitted by Eq. (5) and the coefficient B_0 then can be given. If the current is further increased and goes beyond the breakdown current I_c,[3] then an irreversible and discontinuous change will occur (in Fig. 1, $I_c=9.3$ mA). The nonlinear response described in Fig. 1, which is similar to that of the percolation system in flat substrates,[3] is generally interpreted in terms of a heating and melting process of the hot spots (or links) due to the local Joule heating.[3]

The scaling of I_c as a function of R_0 is shown in Fig. 2. For the lower resistance samples, i.e., $R_0<1$ kΩ, a power-law behavior, $I_c \propto R_0^{-\alpha}$, is observed, which is in agreement with the result of the Pt bilateral rough-film system.[15] From Fig. 2, the critical exponent α is determined to be 0.51 ± 0.05, which is significantly distinct from both the experimental results and the theoretical predictions,[3,9,10,16] providing evidence for the importance of the rough-surface effect.

Several investigations have proved that the exponent α is nonuniversal and it is sensitive to the microgeometry and the materials of both the film and the substrate.[3,9–11] The result above supports this conclusion. An interesting observation is that the α obtained from Fig. 2 is much smaller than 1, which is in contrast with the proposal that $\alpha>1$ was acceptable since the breakdown voltage V_c could not diverge at p_c.[3] However, the result from Fig. 2 is closer to the numerical simulation value on the random fuse model.[9] Therefore, we conclude that the random fuse effect is relatively strong in the Au-film percolation system.

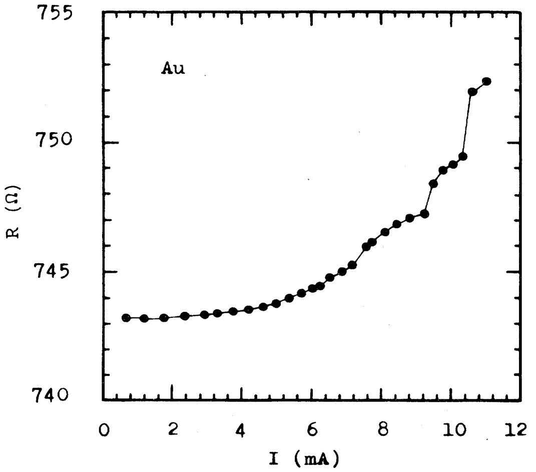

FIG. 1. The R-I behavior of one of the Au bilateral rough samples. The first discontinuity in dR/dI appears at the critical current $I_c=9.3$ mA.

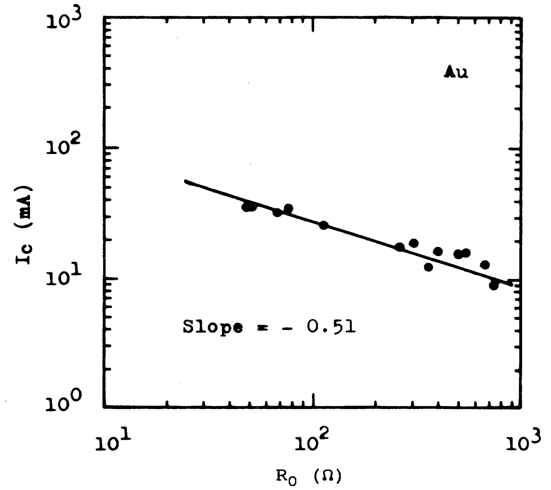

FIG. 2. Scaling of I_c as a function of the Au bilateral rough film resistance R_0.

Since the metallic-film percolation systems generally exhibit the quadratic R-I behavior, i.e., Eq. (5), therefore, it has been proposed that the coefficient B_0 obtained from Eq. (5) could be used for measuring the noise exponent w of the systems.[8,12] In order to experimentally confirm this proposal, we first apply this technique to the Ag-film percolation system deposited onto room-temperature glass substrates by the dc magnetron sputtering method. The coefficients B_0 of the samples with different sheet resistance R_0 are measured and the scaling of B_0 as a function of R_0 is shown in Fig. 3. Obviously, in the Ag system, a power law with the critical exponent $2+w=3.1\pm 0.2$ is well defined up to $R_0=1$ kΩ. This result is in good agreement with the result $2+w=3.2$ obtained by the general ac measurement,[3] indicating that

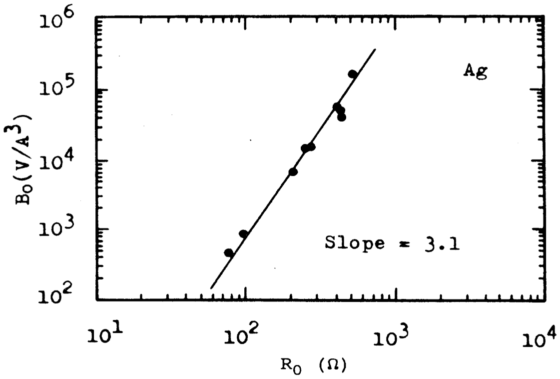

FIG. 3. The scaling of B_0 for the Ag films on flat substrates, $2+w=3.0\pm 0.2$.

the dc method does work in these percolation systems.

As shown in Fig. 4, the Au rough samples also exhibit quadratic *R*-*I* behavior if the current I does not go beyond the critical current I_c. Fitting the experimental data in Fig. 4 by Eq. (5), one finds $B_0 = 5.92\times10^4$ V/A^3. Using this method, the coefficient B_0 of ten Au rough samples with different R_0 are obtained and the scaling behavior is shown in Fig. 5. Again, a power law with the noise exponent $2+w=2.23\pm0.07$ is well defined up to $R_0=1$ kΩ. This value of w is much smaller than the corresponding exponent of Au flat-film system[3,17] and the theoretical predictions in the two-dimensional system.[9,11,18,19] However, it is close to the result of the Pt-film percolation system deposited on the same fractal substrates.[8] The lower noise exponent w has been interpreted as the anomalous hopping and tunneling effects caused by rough surface of the films. It seems that the noise exponent w of the metallic percolation systems deposited on the random fractal substrates $(2<D_0<3)$ are generally smaller than those of the systems on flat substrates. However, to make the final conclusion, further study on various fractal systems is still needed.

Plotting the critical current I_c as a function of the coefficient B_0 yields $I_c \propto B_0^{-x}$ (see Fig. 6). This behavior is similar to that of the flat systems,[3] although their substrates are quite different. From the slope of the experimental data in Fig. 6, one finds $x=0.24\pm0.08$, which is smaller than that of the flat film systems (i.e., $0.36\le x\le0.5$),[3] indicating that the exponent x is strongly dependent on the roughness of the films (or substrates).

It has been proved that, although the noise exponent w is unchanged with the current frequency ω, the third-harmonic coefficient B is intensively independent on ω.[3] In a limited frequency range (i.e., $10<\omega<10^3$ Hz), a power-law dependence $B\propto\omega^{-1/2}$ is observed,[12] which is an indication that the edge effects and the heat capacity of the metallic thin film cannot be neglected.[12] Generally, B_0 is larger than B,[3] then from Eq. (4), one can see that the value of x from Fig. 6 is smaller than that obtained from the ac method is very reasonable. Then the exponent x is actually a function of ω.

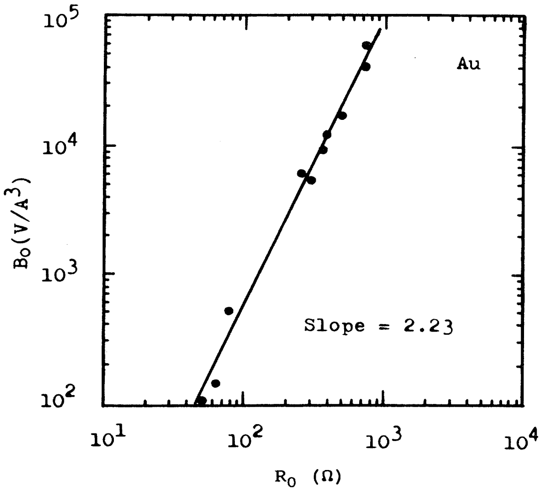

FIG. 5. The scaling of B_0 for the Au rough films, $2+w=2.23\pm0.07$.

Yagil, Deutscher, and Bergman[3] proposed that, resulting from a local melting picture, the exponent x should satisfy

$$\tfrac{1}{2}-1/[2(2t+\kappa)]\le x\le\tfrac{1}{2}\ . \tag{7}$$

In two-dimensional systems, $t=1.3$ and $\kappa=1.1$,[18] hence, they obtain $0.36\le x\le0.5$. Several experimental data supported this proposal. However, these lower and upper

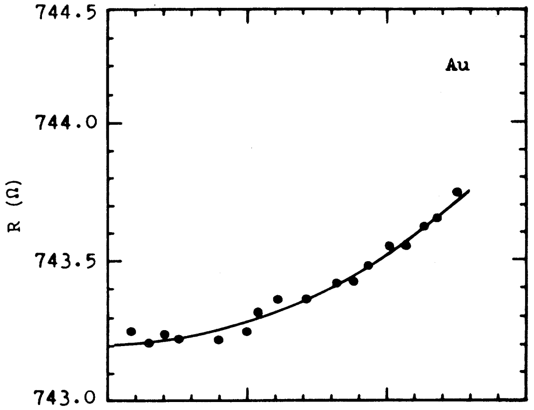

FIG. 4. *R*-*I* characteristic of one of the Au bilateral rough films. This behavior is similar to that of flat-metal films and can be interpreted in terms of the Joule-heating effect. Dots are the experimental data and the solid line represents the fit $R=R_0+B_0I^2$, $B_0=5.92\times10^4$ V/A^3.

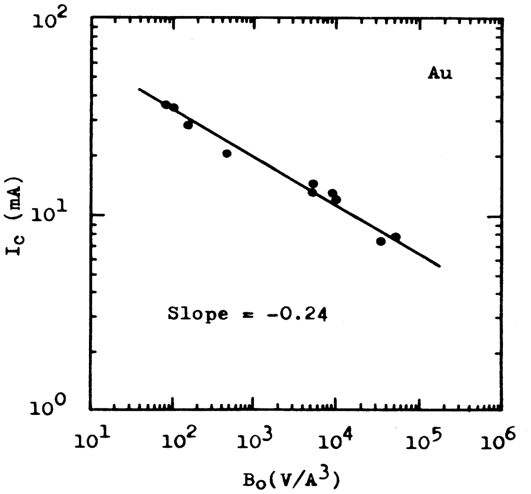

FIG. 6. The scaling of I_c as a function of the coefficient B_0. The straight line describes the power law $I_c\propto B_0^{-x}$, where $x=0.24\pm0.08$ for the Au bilateral rough films.

bounds of x would not be suitable for describing the Au-film samples for two reasons: (1) the strong frequency dependence of B is not taken into account in the derivation of Eq. (7), and (2) in the bilateral rough film system, the dimensionality and microgeometry are closely related to each other,[15,20,21] then both the exponent t and κ would depend on the system dimension, and depend on the microgeometry as well. Therefore, the exact values of t and κ in the bilateral rough system will be quite different from those of the flat film systems. In other words, the physical sense of the universality definition in the bilateral rough-film system is quite confusing. Generally, the dimension and the microgeometry of composites are considered to be two unconcerned quantities. However, when the amplitude of the rough-surface undulate is near or goes beyond the nominal film thickness, there will be a close relation between these two quantities. Then, if the exponent x as well as other exponents depend on the dimension of the system, they are surely sensitive to the microgeometry. Thus, the universal behavior of the system becomes very complicated and the traditional definition of universality[3,22] is improper for the bilateral rough films. In this sense, the bilateral rough film is absolutely a new system and a detailed research on it is quite meaningful for fundamental as well as practical purposes. Therefore, we propose that the discrepancy between the result of Fig. 6 and Eq. (7) can be interpreted by the frequency dependence of B and the bilateral rough-surface effects.

On the other hand, if we assume that $t=1.3$, which has been verified only in two-dimensional systems so far,[3] is still correct in our samples, together with $\kappa = wt$ and the measured value of $w=0.23\pm0.07$ (see Fig. 5), then Eq. (7) gives $0.32 \leq x \leq 0.5$. The lower bound of this inequality is equal to the upper bound of the measured value of x (see Fig. 6). Therefore, we propose that, in the bilateral rough-film system, the decrease of exponent x may be related to the smaller noise exponent w. At present, however, a unified theory of the upper and lower bounds of x for all the percolation systems is still lacking.

It should be noticed that the power-law behaviors generally exist in the Au bilateral rough system (see Figs. 2, 3, 5, and 6). This result supports the proposal that the power-law critical behavior could not be broken by the influence of the fractal substrates.[15] However, a further theoretical demonstration is still needed.

In summary, we have measured the critical current I_c, coefficient B_0, and the relation between I_c and B_0 on a Au-film percolation system deposited on the fractal surfaces of α-Al_2O_3 ceramics. The power-law behavior $I_c \propto R_0^{-\alpha}$ and $I_c \propto B_0^{-x}$, with $\alpha=0.51\pm0.05$ and $x=0.24\pm0.08$, are observed. We propose that the lower critical exponent α can be interpreted as the random fuse effect in the Au rough-film system and the lower exponent x is caused by the frequency dependence of the coefficient B and the relevance between the dimension and microgeometry of the system. The universality definition for the bilateral rough films should be reconsidered since the dimension and microgeometry are no longer irrelevant. This subject remains an interesting topic for further study by both theoretical and experimental methods.

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