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Measurements of the ac conductivity and dielectric constant in a two-dimensional lattice percolation system

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We have fabricated a well-defined two-dimensional lattice percolation system on an aluminum film by using a computer-controlled *x*-*y* plotter. The ac conductivity σ and dielectric constant ϵ of these samples were measured for a frequency range of 10 Hz to 1 MHz. Our data for σ and ϵ near p_c show that the power-law dependences on the stimulating frequency are of the form $\sigma \sim w^x$ and $\epsilon \sim w^{-y}$. Our results ($x = 0.996 \pm 0.050$, $y = 0.024 \pm 0.005$) are in good agreement with previous measurements of a discontinuous gold film prepared by Laibowitz *et al.* However, both results show large discrepancies with the theoretical predictions based on the anomalous diffusion or intercluster polarization effect. Despite these discrepancies, the general scaling law ($x + y = 1$) still holds. We also observed that the intercluster polarization effect plays a more important role than anomalous diffusion in our system.

I. INTRODUCTION

Since Last and Thouless[1] first measured the conductivity in a two-dimensional site percolation system by punching holes in carbon paper, much experimental and theoretical work has been carried out.[2] Due to the enormous difficulties, most of the experiments were carried out on measurements of the dc conductivity.[1,3–5]

The ac response of the percolation system had not been paid much attention until, at the end of 1970's, several theories[6–9] predicted anomalous behavior of the effective σ and ϵ near p_c. Contrary to the flat ac response of each metallic or insulating component, the ac response of the composite of these constituents changes as frequency increases. This phenomenon, dominantly observed near p_c, is called the anomalous ac response. The ϵ and σ, directly related to the complex dielectric constant ($\epsilon_{\text{complex}} = \epsilon + i4\pi\sigma/w$), are not totally independent but connected through causality. So the anomalous response of one quantity could cause the anomalous response of the other.

Bergman and Imry[7] derived a general analytic expression for the effective complex dielectric constant of a random composite as a function of the dielectric constant of each constituent. Their derived frequency dependences for σ and ϵ are $\sigma \sim w^x$ and $\epsilon \sim w^{-y}$ near p_c. The critical exponents x and y satisfy the relation $x + y = 1$. Since this relation can be derived directly even without any scaling assumption, it is called the general scaling relation.

Two different effects, intercluster polarization[6–11] and anomalous diffusion,[12] were considered to explain the anomalous ac response. Intercluster polarization theory emphasizes capacitive coupling between clusters. Throughout the system, and especially near p_c, many conducting paths are blocked off only by thin insulating layers. Since the increase of the area between pure conducting clusters is sharp near p_c, capacitive coupling becomes enormous. This intercluster polarization effect could result in strong nonlinearities in the dielectric response.

This intercluster polarization idea was incorporated with the position space renormalization group[11] or the assumption of scaling.[6,8–10] Both theories give the power law of w with

$$x = t/(t+s), \quad y = s/(t+s) ,$$

where t is the critical-conductivity exponent and s is the dielectric-constant exponent. Since $t = s = 1.3$ for a two-dimensional (2D) percolation system, $x = y = \frac{1}{2}$. Calculated exponents of x and y for three dimensions are 0.72 and 0.28, respectively.

The theory of diffusion on percolation clusters near p_c was extended by Gefen, Aharony, and Alexander[12] for the length scale $a \ll L \ll \xi$. After incorporating the Einstein relation, they obtained the scaling form of the conductivity. Similar, to the intercluster polarization effect, they obtain the power-law behavior of w with $x = t/\nu(2+\theta)$ and $y = (2\nu - \beta)/\nu(2+\theta)$, where $\theta = (t-\beta)/\nu$ is a critical exponent that describes the diffusion on the percolating clusters. In this theory, they consider the delay-time effect due to anomalous diffusion within the clusters. The results obtained are $x = 0.34$ and $y = 0.66$ for two dimensions and $x = 0.58$ and $y = 0.42$ for three dimensions.

The first systematic experiments on the ac response of a percolation system were carried out by Laibowitz and Gefen.[13] By controlling the thickness of a discontinuous gold thin film they could produce a series of percolation systems near p_c. In the critical region, they found the power-law behavior of w. Their measured values of $x = 0.95 \pm 0.05$ and $y = 0.13 \pm 0.05$ were in agreement with the general scaling relation but show large discrepancies with both theories.

By using a carbon-teflon composite, Song *et al.*[14] measured anomalous ac behavior in a three-dimensional system. The measured values of x and y were 0.86 ± 0.06 and 0.12 ± 0.04, respectively. These are closer to the predicted values from the intercluster polarization model than those of the anomalous diffusion theory. Compared to the two-dimensional results, these results are closer to the theoretical predictions. However, the discrepancies still exist. Moreover, the cause of these discrepancies is not fully understood.

In this paper, we report experimental values of σ and ϵ for a two-dimensional lattice percolation system. By using a computer-controlled x-y plotter, we have fabricated a well-defined 2D lattice percolation system on aluminum thin film. The values of σ and ϵ for these samples were measured in the frequency range of 10 Hz to 1 MHz. Since the unit size of bond length ($\sim$mm) is huge, conduction mechanisms other than the intercluster effect or anomalous diffusion are hard to introduce. So electron-electron correlation, hopping, or tunneling conduction could not be included in this system.

II. EXPERIMENTS

When we fabricated the percolating system, we used a heating pen to scribe commercially available aluminum thin film, coated on a polyester sheet.[5] The strong merit of this method is that we can draw the desired pattern by computer.

To observe the anomalous ac response, large capacitance and resistivity are essential for our measurable range of the instrument. To get more capacitive coupling, we chose the modified bond percolation system (Fig. 1). We used two types of aluminum thin films. The thickness of the type-1 sample is almost 1 μm, which is much thicker than that of the type-2 sample. The conductivity of the type-1 sample is almost two orders of magnitude larger than that of type 2. Since the thicknesses of both samples are much less than the unit length or the correlation length of the percolation system, this is truly a two-dimensional percolation system. When we drew insulating lines on type-1 samples, we used high power to cut the thicker films. Thus the combination of the line width and thickness of these two types of sample make their capacitive couplings almost the same. For p near p_c and $p > p_c$, type-2 samples are used because their conductance is within the measurable range for our instrument. Both types of samples were used for $p < p_c$. Since x and y for the conducting region are the same as for the insulating region, and the anomalous ac response appears in a wide range of frequencies for $p < p_c$, it is more profitable to study this anomalous behavior for the insulating region.

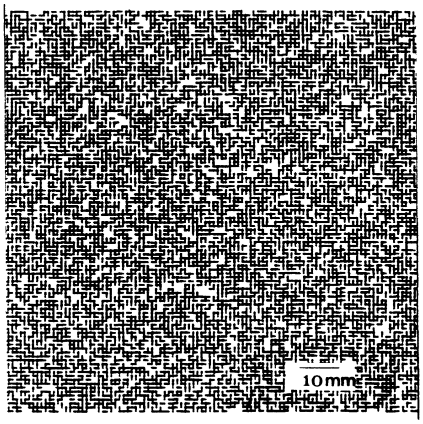

FIG. 1. 100×100 percolation patterns near p_c. The black parts are insulators. The thickness of the black layer shown is slightly thicker than the real one used for our measurements.

We measured the real and imaginary component of σ simultaneously by using a Hewlett-Packard 4192A impedance analyzer with a four-probe geometry. The data acquisition process was controlled by the IBM PC through an IEEE-488 bus cable. To improve the reliability of the data, the signal was averaged over 25 periods. The frequency range of 10 Hz to 1 MHz with 20 frequency points for each decade was chosen for our measurement. To prevent destructive effects in the dielectrics, we applied a low peak to the peak voltage of 0.5 V.

III. RESULTS AND DISCUSSIONS

Since high conductive (type-1) films are not appropriate for examining the ac response in the metallic region, we used the low-conductivity (type-2) films. The conductivity versus frequency is shown in Fig. 2 and the dielectric constant versus frequency in Fig. 3 for samples near p_c. For $p > p_c$, the conductivity is constant in the low-frequency regime, but increases with the power of frequency in the high-frequency region. For $p < p_c$, σ and ϵ are changing for all measurable ranges. The slope in the low-frequency region is smaller than that of the high-frequency region, but the change is not drastic. We obtained the value of x and y, in the best fitting region, by

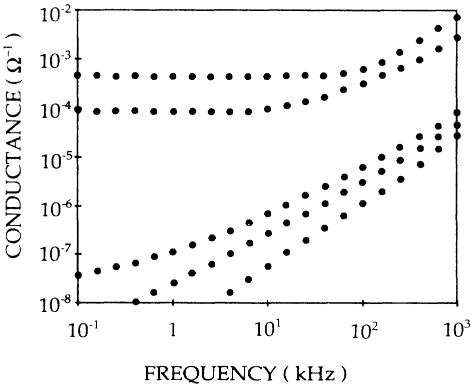

FIG. 2. Conductivity vs frequency for 200×200 (type 2) near p_c.

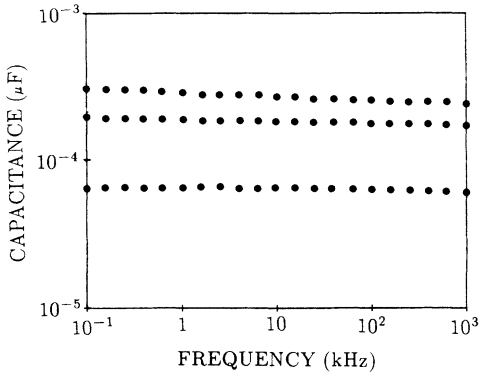

FIG. 3. Capacitance vs frequency for 200×200 (type 2) near p_c.

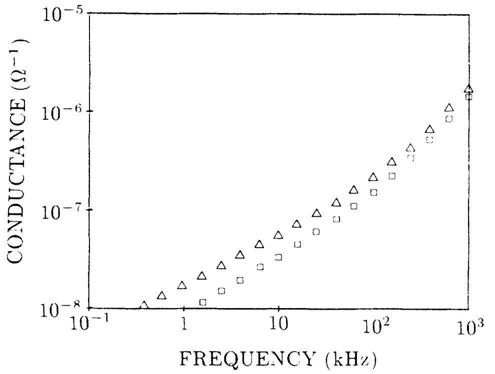

FIG. 5. Conductivity vs frequency for 200×200 (type 1) near p_c (site percolation: square, bond percolation: triangle).

averaging over seven samples just below p_c. The two exponents x and y are 0.996±0.050 and 0.024±0.005, respectively, in good agreement with the results of Laibowitz and Gefen ($x=0.95\pm0.05$, $y=0.13\pm0.05$). Moreover, the general scaling relation is satisfied. Song[15] used an electron-beam lithography technique to make a micron order percolation system, and obtained similar results.

To examine the range of p, where the anomalous behavior occurs, we prepared the samples by making the fill fraction relatively large. The anomalous ac response occurred even in the range of $p-p_c \sim 0.05$, for $p<p_c$. Sample size effects were also investigated, just near p_c, by varying the sample size from 200×200 to 50×50. Size reduction is equivalent to an increase of Δp by the renormalization group, as is clearly seen if we compare curves a and b of Fig. 4. There is no clearly observable change in x and y.

We also carried out ac measurements for several site percolation systems. As far as ac conductivity and the dielectric constant are concerned, theory does not distinguish site and bond percolation. As predicted, the appearance of σ and ϵ is similar to the bond percolation case, and x and y are almost as similar to that case. The general scaling relation is satisfied in the high-frequency region, but not in the low-frequency region, for both systems. (See Figs. 5 and 6.)

This anomalous behavior of the σ and ϵ near p_c might be explained by the following two effects. The first is the coupling between clusters. As we apply higher frequencies, more of the dangling ends contribute to conduction. The other is the delay-time effect of electrons due to diffusion on the fractal structures. The former is more important in two dimensions than in higher dimensions, where electrostatic interaction decreases faster. Since in $d>6$ the polarization of the medium is not singular,[12] anomalous diffusion plays the most important role. These two effects are predicted to appear in the high-frequency regime.

The importance of the intercluster polarization in low dimensions is clearly seen in our measurements, as follows. If we heat the scribing pen more, the thickness of the insulating layer increases and capacitive coupling becomes weak. Since the anomalous ac response is not well observed for the thicker insulating layered samples, and anomalous diffusion theory is independent of the capacitive coupling, anomalous diffusion is not the major mechanism for this anomalous ac response. From the above

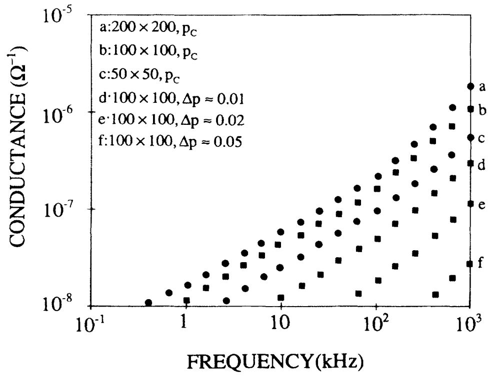

FIG. 4. The ac conductivity vs frequency for different sample sizes at p_c and for different p with 100×100 size.

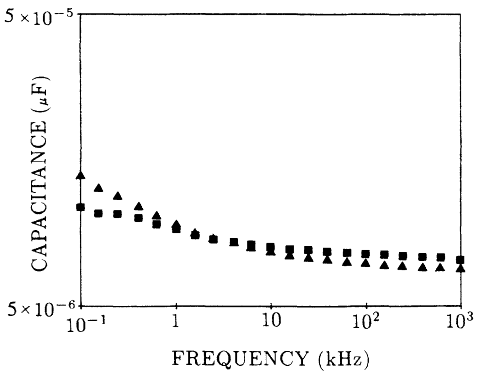

FIG. 6. Capacitance vs frequency for 200×200 (type 1) near p_c (site percolation: square, bond percolation: triangle).

results, we conclude that the intercluster polarization effect is much more important than anomalous diffusion effects in our two-dimensional experiment.

IV. CONCLUSIONS

We have measured the anomalous ac response in the well-defined 2D lattice percolation system. The observed power-law behavior of σ and ϵ was as predicted by the intercluster polarization and anomalous diffusion theories. However, the observed critical exponents $x=0.996\pm0.050$ and $y=0.024\pm0.005$ do not fit the predicted values of either theory, though the general scaling relation is still valid. By changing the capacitance coupling, we observed that the intercluster polarization effect plays a more important role than anomalous diffusion in our system.

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