Temperature-dependent ac conductivity of thin percolation films

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We report complex ac-conductivity $\sigma(\omega)$ measurements on thin-gold-film percolation networks at temperatures between 300 and 4.2 K and at frequencies between 100 Hz and 1 GHz. $\sigma(\omega)$ is highly temperature dependent below 100 K. A transition between a high-temperature state exhibiting scaling in $\sigma(\omega)$ and a low-temperature state exhibiting unusual frequency dependencies occurs near 35 K. This transition temperature appears independent of the parameters which characterize the fractal nature of these films. In contrast to the high-temperature behavior, the low-temperature state cannot be accounted for by a conventional scaling theory of percolation.

I. INTRODUCTION

The field of percolation physics, with its close association with fractal geometry, has been actively studied during the past decade.[1] The percolation problem is concerned with the physics of dilute systems which are in the vicinity of their critical points. Near the critical point the system is characterized by a site-filling probability p which is close to the percolation threshold probability p_c. For values of p less than p_c, isolated "clusters" form which do not allow complete percolation to occur across the entire system. For example, in a metal-insulator mixture, the dc electrical conductivity $\sigma_{dc}=0$ for $p<p_c$. With $p>p_c$, a network of metal spans the specimen and $\sigma_{dc}\neq 0$.

The scaling theory of percolation is concerned with the way in which physical properties of a system (such as the electrical conductivity) scale with the site-filling probability.[2] Properties scale as $|p-p_c|^{\alpha}$, where the critical exponent can be positive or negative depending upon the property in question. Although p_c may depend sensitively on the details of the system or underlying lattice, the critical exponents are *universal* in that they are theoretically independent of the details of the underlying lattice, and depend solely on the *dimensionality* of the system involved. Many of the properties of percolation systems thus follow universal scaling laws. The universal behavior of percolation systems has been extensively studied by computer simulations[3-5] and experiments performed on both fabricated and natural percolation networks.[6-8]

Scaling theory predicts that, in a self-similar, fractal metal-insulator percolation network, σ_{dc} scales as[2,9]

$$\sigma_{dc}\sim(p-p_c)^{\mu}\sim\xi^{-\mu/\nu}, \tag{1}$$

where $\xi\sim|p-p_c|^{-\nu}$ is the percolation correlation length. This result holds for $p>p_c$. The self-similar nature of the percolation network also gives rise to scaling between the complex ac conductivity and the applied frequency. Theories which account for the anomalous diffusion on the fractal network suggest that the resistance R and capacitance C should scale with frequency ω as[2]

$$R\sim\omega^{-x} \tag{2}$$

and

$$C\sim\omega^{-y}, \tag{3}$$

where $x+y=1$. This scaling holds for frequencies where the anomalous diffusion length L_d falls within the range $a_0\ll L_d\ll\xi$, where a_0 is the microscopic lattice size.

Studies of extremely thin metal-insulator films[8] indicate that they belong to the same universal class as the idealized percolation systems. The films are fabricated by evaporating gold atoms onto an insulating substrate in such a way that a complete coverage of metal does not occur. Digitized transmission electron micrographs[10] show that such films are geometrically equivalent to an idealized percolation system, indicating that the results of scaling theory should apply to their physical properties.

Laibowitz *et al.*[11] have measured the room-temperature complex conductivity $\sigma(\omega)$ on gold fractal films at frequencies up to 10 MHz. $R=1/\mathrm{Re}\sigma(\omega)$ and $C=\mathrm{Im}\sigma(\omega)/\omega$ were observed to scale with frequency in qualitative agreement with Eqs. (2) and (3), although the critical exponents (x and y) were found to be in disagreement with those expected. This discrepancy has been attributed to the "nonideal" nature of the system, perhaps arising from intercluster effects in the form of electron-electron interactions. Such interactions, which may be frequency and temperature dependent, are not accounted for in conventional scaling theory.

We have extended the measurement of $\sigma(\omega)$ on gold fractal films to frequencies between 100 Hz and 1 GHz, and to temperatures between 300 and 4.2 K. $\sigma(\omega)$ is found to be very temperature dependent below 100 K. A "transition" from a high-temperature states which displays scaling ac conductivity to a low-temperature state which displays highly modified scaling behavior occurs at roughly 35 K. This transition temperature appears to be insensitive to the parameters which characterize the fractal nature of the films; this indicates that the transition may arise from the underlying physical attributes of the films which are not accounted for in the scaling theory.

A description of our percolation films and the tech-

niques used to measure $\sigma(\omega)$ are presented in Sec. II, together with experimental $\sigma(\omega)$ data. The data are analyzed and a brief description of the scaling theory of ac conductivity is given in Sec. III. In Sec. IV we summarize our findings and discuss the direction of future research.

II. EXPERIMENT

The gold films used in this study were fabricated at the IBM Thomas J. Watson Research Center in Yorktown Heights, NY. A complete description of the fabrication technique is given in Ref. 8. The specimens consist of extremely thin gold films that have been evaporated onto insulating substrates. The nominal coverage of the films is very close to the percolation threshold coverage, with a typical average film thickness of 6–10 nm. The films are percolation systems with dc resistances in the range of 10 Ω to 1 MΩ. Previous transmission-electron-microscopy (TEM) work[10] on similar films suggests that they are geometrically analogous to a percolation system, with a typical microscopic lattice size of $a_0 \approx 10$ nm, percolation correlation lengths on the order of 100 nm, and a fractal dimension of $D \approx 1.90$.

In the study reported here, the ac conductivity was measured in the frequency range 100 Hz to 1 GHz using two different methods. In the low-frequency range (100 Hz to 10 MHz) a Hewlett-Packard HP 4192A low-frequency impedance analyzer, functionally equivalent to a lock-in detector, was used. In the high-frequency range (4 MHz to 1 GHz) a computer-controlled HP 8754A network analyzer was employed. In this high-frequency–measurement system, the sample forms the termination of a rigid 50-Ω coaxial cable. A rf electromagnetic wave was transmitted down the cable, and the reflected wave detected. The reflection coefficient Γ was corrected via a three-point (open, 50 Ω, short) calibration technique; this correction also removed any phase offset due to electrical line length. The ac conductivity is determined from the corrected reflection coefficient via the relation

$$Z = \frac{1+\Gamma}{1-\Gamma} , \tag{4}$$

where Z is the normalized impedance. The resistance is the real part of the impedance ($Z = R + iX$), while the capacitance is related to the imaginary part of the conductance ($Z^{-1} = G + iB$) by $C = B/\omega$. Both methods used to measure $\sigma(\omega)$ provided very accurate resistance data at all frequencies between 10 Hz and 1 GHz. Because the instruments directly measure the conductivity ($B = C\omega$) rather than the capacitance, the capacitance could not be determined accurately at very low frequencies. Hence, we report capacitance data only in the high-frequency regime, starting at 10–100 kHz, with a sensitivity of roughly ±0.1 pF.

In both the high- and low-frequency regimes, care was taken to ensure that a low signal level was applied to the sample to avoid heating. The samples were mounted to the end of the coaxial cable with short 1-mil gold leads, using conductive silver paint. The samples were cooled in a helium-gas–flow cryostat.

The ac conductivity $\sigma(\omega)$ of a gold percolation film with a room-temperature dc resistance of 380 Ω is shown in Figs. 1–5. Both the resistance R and capacitance C are essentially unaffected by changes in temperature above 100 K. The frequency-dependent R and C at both 300 and 100 K are presented in Fig. 1. The data are nearly identical at both temperatures. Similar measurements were made at 50-K intervals between 300 and 100 K, and no change in the frequency-dependent behavior was observed. R is independent of frequency below a critical frequency of $\omega_\xi \approx 300$ kHz, with a low-frequency value of $R(\omega < \omega_\xi) = 380$ Ω. Similarly, C appears independent of frequency below roughly 2 MHz, with a low-frequency value of $C_0 = 200$ pF. Above these critical frequencies, both R and C decreases with increasing frequency. At 1 GHz, R and C have dropped to 25 Ω and 20 pF, respectively.

When fitted to the simple scaling-frequency expressions as given in Eqs. (2) and (3), the high-frequency behavior of R and C from Fig. 1 gives values for the critical exponents of $x = 0.35 \pm 0.05$ and $y = 0.71 \pm 0.05$. Hence, above 100 K this gold percolation film displays the expected[2] frequency-scaling ac conductivity with no apparent temperature effects on either the overall scaling behavior, the low-frequency resistance, or the low-frequency capacitance. In addition, there do not appear to be any effects of temperature on the critical frequencies ω_ξ above which the the ac scaling behavior occurs.

In contrast to the high-temperature state where $\sigma(\omega)$ is temperature independent, below 100 K $\sigma(\omega)$ becomes strongly temperature dependent. Figure 2 shows $R(\omega)$ for selected temperatures below 100 K. The relative change of R above ω_ξ decreases with decreasing temperature. This is most evident in the data at 23 K, where R is constant at all frequencies up to 1 GHz. While the high-frequency scaling behavior appears to diminish with

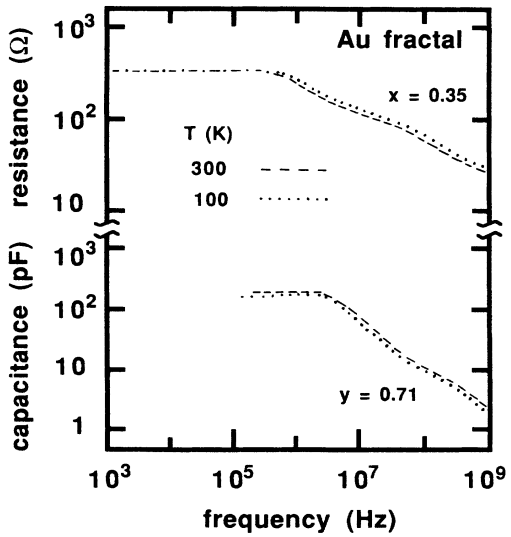

FIG. 1. Resistance and capacitance data at both 300 K (dashed lines) and 100 K (dotted lines). The scaling exponents x ($R \sim \omega^{-x}$) and y ($C \sim \omega^{-y}$) are indicated in the figure.

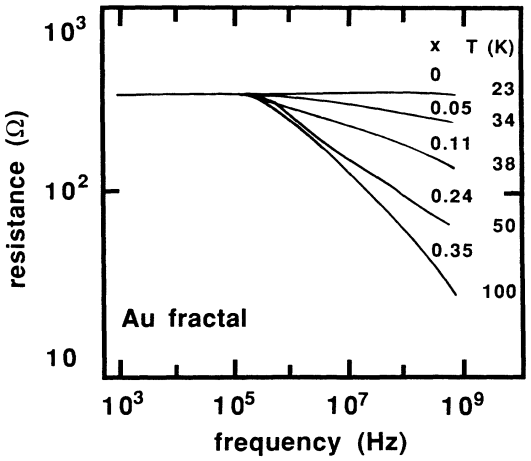

FIG. 2. Resistance data in the vicinity of T_c. The critical exponents $x(T)$ are indicated in the figure.

dropping temperature, R below the critical scaling frequency is unchanged by a reduction in temperature. In addition, the critical frequency remains independent of temperature. Hence, only the high-frequency ac resistance above ω_ξ appears temperature dependent. Below 23 K the percolation film shows no frequency-scaling behavior, acting instead like a homogeneous metal. In terms of scaling theory it would appear that the critical exponent x, which controls the resistance at frequencies above ω_ξ, drops to zero below 23 K.

The capacitance of the gold percolation film below 100 K is shown in Fig. 3. The data indicate that C is also very temperature dependent below 100 K. Between 50 and 23 K, C drops by roughly 3 orders of magnitude, but still appears to scale with frequency. The critical capacitance exponent appears to reach a low-temperature limit of $y=1$. At 23 K, C is less than the experimental sensitivity of 0.1 pF at all frequencies. As a result, we are not able to determine if C scales with ω below 23 K.

Figures 4(a) and 4(b) show, respectively, the detailed temperature dependence of $R(\omega)$ and $C(\omega)$ over a wide temperature range. The data clearly suggest that a "transition" occurs in the percolation film somewhere below 50 K. Figure 5(a) and 5(b) show $R(\omega)$ and $C(\omega)$ near the transition region. The effects of the transition begin to appear in the frequency-dependent conductivity data at roughly 50 K, and the transition appears to be complete below 25 K. Hence, we identify a critical transition temperature $T_c \approx 35$ K, and a width T_w of approximately 25 K.

Measurements on a number of other gold percolation samples show the same general temperature-dependent $\sigma(\omega)$ behavior with transition temperatures ranging from 35 to 40 K. The dc resistances of the samples studied ranged over 4 orders of magnitude, from 90 Ω to 500 kΩ, with the 500-kΩ sample being very close to the metal-insulator percolation threshold. Because the dc resistance scales with the percolation correlation length, these samples span a wide range of percolation parameters, from being very metallic ($p \approx 1$) to being very nearly insulating ($p \approx p_c$). In all cases the temperature-dependent $\sigma(\omega)$ of the films were found to obey the same general be-

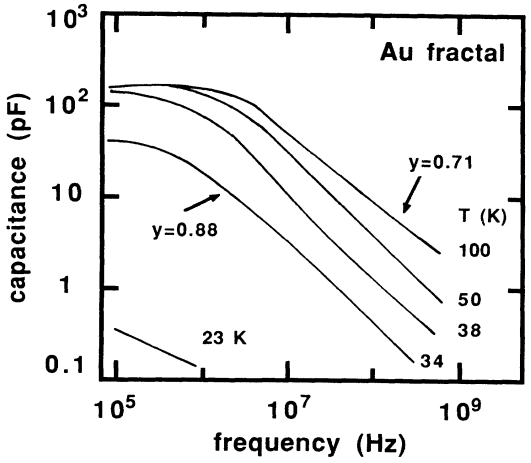

FIG. 3. Capacitance data in the vicinity of T_c. The critical exponents $y(T)$ are indicated in the figure.

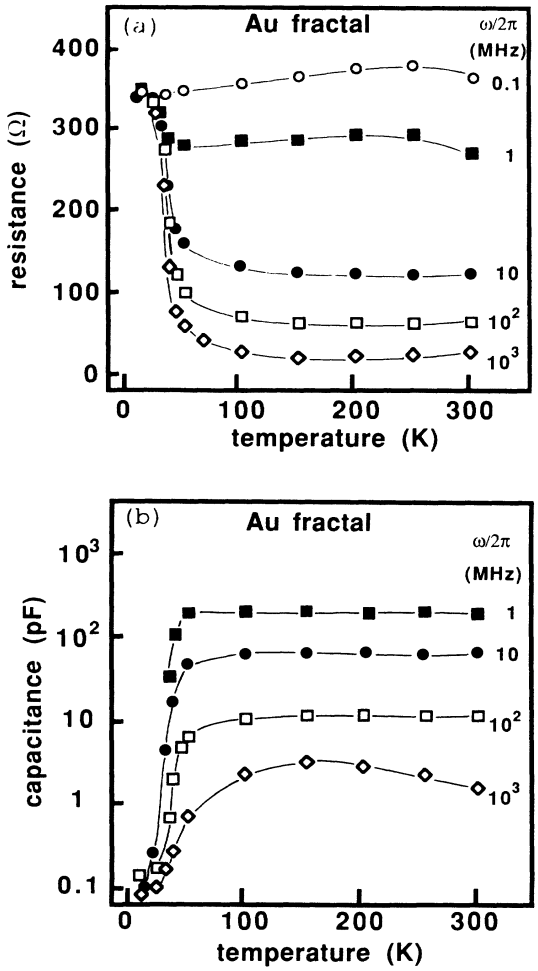

FIG. 4. (a) $R(\omega)$ and (b) $C(\omega)$ plotted as a function of temperature below 300 K. The solid lines are intended as guides to the eye.

havior shown in Figs. 4 and 5 with a transition near 35–40 K separating a high-temperature state which displays scaling in $\sigma(\omega)$ and a low-temperature state which exhibits no scaling in $\sigma(\omega)$. Both the dc resistance and the critical frequency (ω_ξ) beyond which scaling behavior occurs were found to be temperature independent for each film.

Figure 6 shows the transition width T_w and the critical frequency ω_ξ for different films as a function of room-temperature dc film resistance. More resistive films (p closer to p_c) have broader transitions, while ω_ξ decreases dramatically with increasing film resistance. A strong correlation thus exists between the dc resistance, and hence the percolation correlation length, and ω_ξ. The transition width T_w appears to saturate at 40 K for film resistances $R_{dc} > 1$ kΩ; hence, T_w does not scale with film resistance in as simple a form as does ω_ξ.

In comparing $\sigma(\omega)$ for different films, the following trends are observed. Although R_{dc} varied by as much as 4 orders of magnitude, the low-frequency ($\omega \ll \omega_\xi$) film capacitance was on the order of 200 pF for all films. Hence, $C(\omega \to 0)$ is independent of $|p - p_c|$ for the range of films considered here. Furthermore, the frequency-dependent conductivity in high-resistance films suggests that there are *two* frequency-scaling regimes for films with small ω_ξ. This is evident from Fig. 7, which shows $\sigma(\omega)$ measured at room temperature for a film with $R_{dc} = 2.2$ kΩ and $\omega_\xi = 60$ kHz. At frequencies between ω_ξ and 3 MHz, the resistance scales with a critical exponent of $x = 1 \pm 0.05$, while the capacitance does not scale with frequency ($y = 0$). Above 3 MHz the resistance exponent drops to $x = 0.32 \pm 0.05$ and the capacitance exponent rises to $y = 0.8 \pm 0.05$. In both frequency regimes the sum of the critical exponents is always one within the experimental uncertainty.

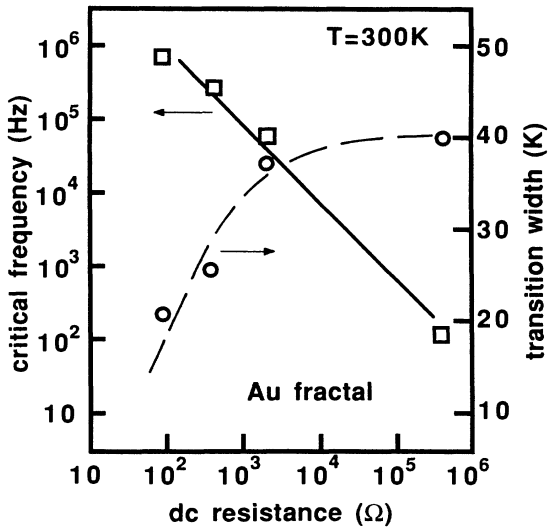

FIG. 6. Plots of critical frequency (squares) and transition width (circles) as a function of dc resistance. The dashed line is a guide to the eye. The straight line is a fit to $\omega_\xi \sim R_{dc}^{-0.82}$.

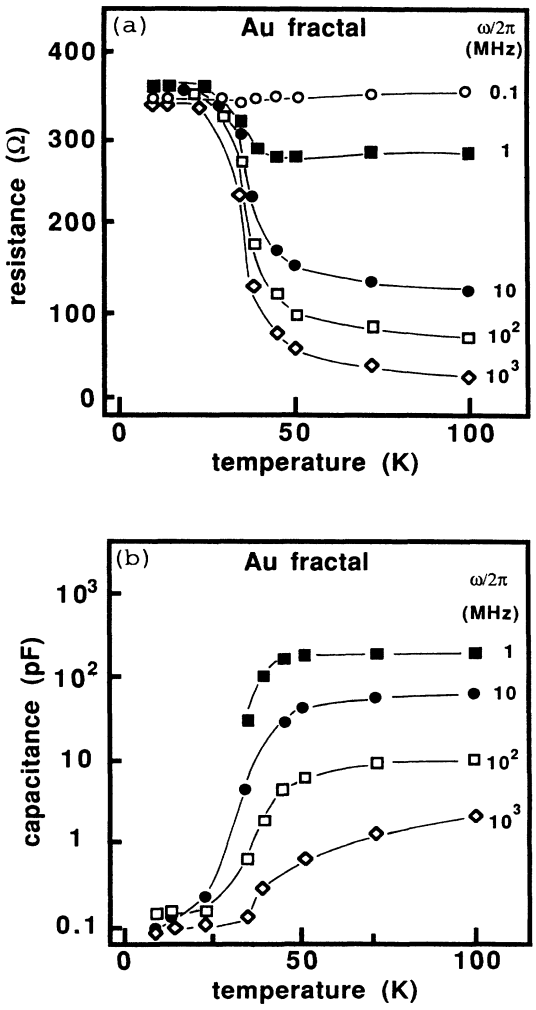

FIG. 5. (a) $R(\omega)$ and (b) $C(\omega)$ near the transition region. The solid lines are intended as guides to the eye.

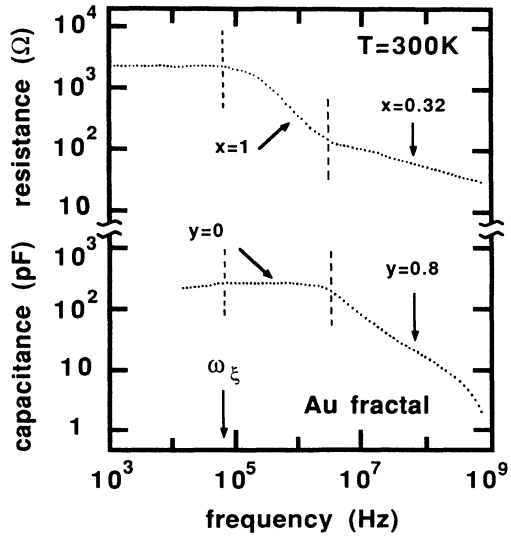

FIG. 7. Room-temperature conductivity of a Au film with a larger dc resistance. The resistance and capacitance data are plotted as a function of frequency. The critical exponents x and y are as indicated in the medium- (60 kHz $< \omega/2\pi <$ 3 MHz) and high-frequency ($\omega/2\pi >$ 3 MHz) regimes.

III. ANALYSIS

A. Scaling theory

We first examine in some detail the theory which predicts that a scaling relationship should exist between the macroscopic properties of a percolation system and its underlying microscopic structure. The theory is based on the premise that the many physical properties of a percolation system will scale with the site-filling probability p in the form $|p-p_c|^{\alpha}$, where α is the critical exponent. Critical exponents that relate to the work here are ν, the correlation-length exponent; μ, the dc-conductivity exponent; and β, the infinite network lattice exponent. The exponent ν determines the percolation correlation length, given by

$$\xi \sim |p-p_c|^{-\nu}, \tag{5}$$

where ξ is measured in units of the microscopic lattice length a_0. Hence, the correlation length is actually $a_0\xi$. Previous TEM studies[10] indicate that gold percolation films have a microscopic lattice size $a_0 \approx 10$ nm and a correlation length of roughly 100 nm, yielding $\xi \approx 10$. Computer simulations on two- and three-dimensional percolation networks suggest $\nu_{2D}=1.35$ and $\nu_{3D}=0.88$.[2,12]

For $p > p_c$ the critical exponent β determines the probability for a point in the lattice to belong to the infinite network, $P_\infty(p)$. This probability is given by

$$P_\infty(p) \sim (p-p_c)^{\beta} \sim \xi^{-\beta/\nu}. \tag{6}$$

the low-frequency network capacitance C_0 is also related to β by

$$C_0 \sim \xi^{2-\beta/\nu}. \tag{7}$$

Computer simulations yield $\beta_{2D}=0.14$ and $\beta_{3D}=0.4$.[2] The dc conductivity depends on the exponents μ and ν through the expression

$$\sigma_{dc} \sim (p-p_c)^{\mu} \sim \xi^{-\mu/\nu}.$$

Computer simulations suggest $\mu_{2D}=1.2$ and $\mu_{3D}=2.0$.[2,12] The low-frequency capacitance and resistance are clearly tied to the percolation correlation length. As a result, the scaling theory directly relates the behavior of R and C at high frequencies to the underlying nature of the percolation system through ξ.

These low-frequency results represent averages taken over length scales L_s much larger than ξ. On these length scales the percolation film appears homogeneous. As the length scale over which the system is examined is reduced below ξ, its inhomogeneous nature becomes evident. Thus, when measuring the resistance at $L_s > \xi$ the film resistance should take on a constant value which reflects the microscopic nature of the film (this resistance can be quite high because the infinite cluster is extremely thin). In the opposite extreme, with $L_s \approx a_0$, the gold film should act like a very perfect metal, because on this length scale the measurement probes the gold atoms directly. For $a_0 < L_s < \xi$ the fractal nature of the film will become less evident and the resistance should drop as L_s approaches a_0.

An ac signal at a frequency ω will measure the conductivity over a length $L(\omega) \sim 1/\omega$. Hence, at low frequencies, where $L(\omega) \gg \xi$, the resistance should be independent of frequency. Above a critical frequency ω_ξ, where $L(\omega_\xi) \approx \xi$, the signal will begin to probe within the percolation cluster and the resistance should drop. The capacitance is expected to behave in a similar fashion. At $\omega < \omega_\xi$ the intercluster capacitance will give rise to a nonzero total capacitance. As the frequency rises above ω_ξ, the signal probes more directly the metallic constituents which form the percolation network, and the capacitance should therefore drop.

Two separate methods have been employed to characterize the high-frequency behavior of the resistance and capacitance in percolation systems.[9,13–16] Although the two theories are based on different starting assumptions, they both predict that the resistance and capacitance should scale with frequency in the form

$$R \sim \omega^{-x}$$

and

$$C \sim \omega^{-y}.$$

The two theories differ only in their expressions for x and y. In the *noninteracting cluster* approximation it is assumed that anomalous diffusion within a percolation cluster occurs.[9,13] The anomalous diffusion comes about because of the fractal nature of the infinite percolation network. When performing a random walk on the percolation backbone, the mean-square distance traveled will scale with the travel time t in the form

$$\langle r^2(t) \rangle \sim t^{2/(2+\theta)}, \tag{8}$$

where θ is related to the critical exponents via $\theta=(\mu-\beta)/\nu$. The θ term in Eq. (8) results from the self-similar nature of the infinite cluster; for a nonfractal system, $\theta=0$, and the mean-square displacement scales linearly with t, as expected for a nonfractal, Euclidean system. In two dimensions the values previously quoted for the critical exponents give $\theta=0.79$. By performing a Fourier transform and replacing the time in Eq. (8) with frequency ($t \Longrightarrow 1/\omega$), the frequency-dependent length scale $L(\omega)$ becomes

$$L(\omega) \sim \omega^{-1/(2+\theta)}. \tag{9}$$

The conductivity is calculated by relating this diffusion length to σ via the Einstein diffusion relation. In the noninteracting assumption the critical exponents of the frequency-dependent resistance and capacitance become[8,12]

$$x = \frac{\mu}{\nu(2+\theta)} = \frac{\mu}{2\nu-\beta+\mu} \tag{10}$$

and

$$y = \frac{2\nu-\beta}{\nu(2+\theta)} = \frac{2\nu-\beta}{2\nu-\beta+\mu}. \tag{11}$$

These exponents sum to 1 ($x+y=1$) because the frequency dependence of both the resistance and capaci-

tance are characterized by the same time scale.[9,11] These exponents hold in the critical frequency regime where $L(\omega) > \xi$. From Eq. (9) the critical frequency is given by

$$\omega_\xi \sim \xi^{-(2+\theta)} \sim \xi^{-(2\nu+\mu-\beta)/\nu} . \quad (12)$$

In two dimensions the values for μ, ν, and β suggest that the scaling exponents should be $x = 0.32$ and $y = 0.68$.

The *intercluster polarization* approach assumes that scaling conductivity arises solely from intercluster effects.[14,16] In this model the neighboring clusters are linked together with an interaction capacitance. As a result, the critical resistance exponent x is unchanged from the previous result [Eq. (10)], while the critical capacitance exponent becomes

$$y = \frac{s}{\nu(2+\theta)} = \frac{s}{2\nu-\beta+\mu} , \quad (13)$$

where s is a critical exponent which characterizes the polarizability of the system. The two expressions for y [Eq. (11) and (13)] are equivalent only if $s = 2\nu - \beta$. It is theoretically unclear if this equality holds true in all dimensions,[9,17] although it has been suggested that the equality $s = \mu$ may be true in two dimension.[18–20] With the accepted values for the critical exponents, this would give $x = y = 0.32$, which is in disagreement with the requirement that x and y sum to 1. By requiring both that $x + y = 1$ and $\mu = s$, x and y become $x = y = 0.5$. Clearly, measurements of x and y in real systems will indicate which scaling approach is more correct.

B. High-temperature scaling behavior

We now compare the high-temperature ac-conductivity measurements to these scaling theories. The results presented in this work agree with the scaling theories in that the resistances and capacitances do scale with frequency past a critical frequency which scales inversely with the dc resistance. Throughout the critical frequency region, the critical exponents do roughly sum to 1 (experimentally, $x + y \approx 1.0 \pm 0.2$). Above 1 MHz the critical exponents are roughly given by $x \approx 0.3 \pm 0.05$ and $y \approx 0.7 \pm 0.05$, in excellent agreement with the noninteracting cluster assumption. As the data in Fig. 7 indicate, films with $\omega_\xi < 1$ MHz also display an intermediate-frequency region where $x \approx 1$ and $y \approx 0$, in reasonable agreement with the interacting cluster assumption. This compares favorably with earlier low-frequency measurements that were performed on these films.[11] That work found $x \approx 0.95 \pm 0.05$ and $y \approx 0.13 \pm 0.05$ below 10 MHz. Thus, it would seem that intercluster effects appear important at intermediate frequencies ($\omega_\xi < \omega < \sim 1$ MHz), while anomalous diffusion is more important at high frequencies. A scaling theory which unifies these two scaling mechanisms has not yet been developed.

Scaling theory makes a number of other predictions about the physical properties of these films. ω_ξ should inversely scale with R_{dc}, and this is confirmed by the data shown in Fig. 6. While the general scaling relationship appears to hold, the measured exponent (-0.82) disagrees with the predicted exponent (-1.66). Scaling theory also cannot account for the fact that the low-frequency capacitance appears to be independent of R_{dc}. Thus, although the predictions of the scaling theory are borne out to some extent by these results, it cannot account for all aspects of the high-temperature data.

C. Low-temperature behavior

We now discuss the low-temperature results. Below 100 K the resistance and capacitance become very temperature dependent. A transition from a high-temperature state displaying scaling ac conductivity ($x, y \neq 0$) to a low-temperature state displaying no apparent scaling behavior occurs near $T_c \approx 35$ K. This low-temperature state is characterized by frequency-independent resistance ($x = 0$) and zero capacitance (C is certainly less than 0.1 pF). While x and C are very temperature dependent, the dc resistance and the critical frequency remain unaltered by changes in temperature. It would appear that these results cannot be accounted for by conventional scaling theory.

Near T_c it appears that scaling may still occur in these films. In the temperature range near T_c ($T_c - T_w/2 < T < T_c + T_w/2$), the resistance exponent drops to zero while the capacitance exponent y appears to rise to 1. Hence, $x + y \approx 1$ in this temperature regime. This suggests that scaling may still occur in the low-temperature state, and it is not observable there only because the capacitance is no longer measurable ($C < 0.1$ pF).

In order for the scaling theory to account for this low-temperature behavior, the critical exponents μ, ν, and β must be altered near and below T_c. The theory can give the proper conductivity exponents ($x = 0$, $y = 1$) if we assume $\mu = 0$. While the changes in x and y can be accounted for in this way, scaling theory would then suggest that ω_ξ, R_{dc}, and C_0 should change in ways that are inconsistent with the data. Specifically, with $\mu \Longrightarrow 0$, R_{dc} should decrease, ω_ξ should increase, and C_0 should stay unchanged. In actuality, both R_{dc} and ω_ξ are independent of temperature, while C_0 drops from 200 pF at 100 K to < 0.1 pF at 23 K. According to Eq. (12), the theory can force ω_ξ to be constant if we assume both that μ goes to zero and β becomes $\beta - \mu$, but R_{dc} should then drop to zero and C_0 should increase, contrary to experimental observations. Thus, the theory of frequency-scaling ac conductivity breaks down at low temperatures. This is somewhat surprising in light of the fact that the theory does not consider temperature effects, and from this it might be assumed that it should work best in the low-temperature limit. In actuality, it appears that scaling theory applies to our gold fractal films only in the high-temperature limit.

It is instructive to consider possible failures of scaling theory as applied to our gold films at low temperatures. First, Figs. 2 and 3 show that at low temperatures the complex conductivity $\sigma(\omega)$ becomes independent of frequency over the entire measured frequency range ($\mathrm{Re}\sigma \sim 1/R$, independent of ω in Fig. 2, and $\mathrm{Im}\sigma \sim \omega C$, independent of ω in Fig. 3). Hence there is no extra contribution to the ac conductivity at higher frequency, as expected for a metal but not a percolation network. On the other hand, the dc conductivity is temperature in-

dependent at low temperatures, in contrast to good metallic behavior. Hence the transition near 35 K does not mark a simple percolation-to-metal transition; rather, it marks a transition from a good percolation network to a state where other effects dominate, possibly intracluster surface scattering.

It is also important to note that the scaling theory was originally intended to model the behavior of abstract percolation systems that are "created" in computer simulations. In these simulations the clusters consist of groups of nonphysical points lying in a two-dimensional lattice. In the gold films these clusters consist of mounds of gold atoms. Hence, the films will not be entirely two dimensional. Our films have thicknesses that range from 6 to 10 nm, while the percolation correlation lengths are roughly 100 nm. Strictly speaking, the clusters are (microscopically) three-dimensional objects. In addition, the gold atoms congregate together within the clusters in an ordered manner. Temperature effects will also cause phonons to exist throughout the film. There may also be interactions between the substrate and the gold which may be temperature dependent [we, however, note that the temperature behavior of $\sigma(\omega)$ cannot be ascribed simply to thermal contractions of the substrate, which might occur with dropping temperature, because this would also necessitate changes in R_{dc} and ω_{ξ}, none of which are borne out in the data]. All of these many physical aspects of the gold films are not considered in the scaling theory. While these underlying physical attributes may not be important when studying the film's structural nature (TEM work show these films to appear self-similar in exactly the same way that simulated percolation networks do),[10] they may have a profound influence upon the film's dynamical behavior. Hence, the physical, "nonideal" nature of these gold percolation films may account for the highly temperature-dependent behavior exhibited in $\sigma(\omega)$ below 100 K. On the other hand, the fact that some scaling ($x = 0$, $y = 1$) still occurs near T_c suggests that a simple extension of conventional scaling theory may account for the experimental observations made at low temperatures in these real systems.

IV. CONCLUSIONS

The temperature-dependent ac conductivity of these gold percolation films show that the traditional scaling theory breaks down at low temperatures. In the high-temperature regime the theory is in accord with the measured data, indicating that anomalous diffusion gives rise to scaling ac conductivity above 10 MHz. Below 100 K the resistance and capacitance no longer show conventional scaling behavior. This cannot be accounted for by any of the scaling theories which attempt to describe percolation systems. The fabrication and study of percolation films made from materials different from those used here could give insights into the exact nature of the physical film properties which control the low-temperature behavior.

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[1]For a review, see S. H. Liu (unpublished).
[2]D. Stauffer, Phys. Rep. **54**, 3 (1979); S. Halvin and D. Ben-Avraham, Adv. Phys. **36**, 695 (1987).
[3]H. E. Stanley, J. Phys. A **10**, L211 (1977).
[4]P. L. Leath, Phys. Rev. B **14**, 5046 (1976).
[5]R. J. Harrison, G. H. Bishop, and G. D. Quinn, J. Stat. Phys. **19**, 53 (1978).
[6]B. A. Ables, H. L. Pinsh, and J. I. Gittleman, Phys. Rev. Lett. **35**, 247 (1976).
[7]C. J. Lobb, M. Tinkham, and W. J. Skoçpol, Solid State Commun. **27**, 1253 (1978).
[8]R. B. Laibowitz, E. I. Allessandrini, C. R. Guarnieri, and R. F. Voss, J. Vac. Sci. Technol. A **1**, 438 (1983).
[9]Y. Gefen, A. Aharony, and S. Alexander, Phys. Rev. Lett. **50**, 77 (1983).
[10]R. F. Voss, R. B. Laibowitz, and E. I. Allessandrini, Phys. Rev. Lett. **49**, 1441 (1982).
[11]R. B. Laibowitz and Y. Gefen, Phys. Rev. Lett. **53**, 380 (1984).
[12]R. B. Pandey and D. Stauffer, Phys. Rev. Lett. **51**, 527 (1982).
[13]D. Ben-Avraham and S. Harlin, J. Phys. A **15**, L691 (1982).
[14]A. L. Efros and B. I. Shklovski, Phys. Status Solidi B **76**, 475 (1976).
[15]J. P. Straley, Phys. Rev. B **15**, 5733 (1977).
[16]D. J. Bergmann and Y. Imry, Phys. Rev. Lett. **39**, 1222 (1977).
[17]M. J. Stephen, Phys. Rev. B **17**, 4444 (1978).
[18]A M. Dykhne, Zh. Eksp. Teor. Fiz. **59**, 110 (1970) [Sov. Phys.—JETP **32**, 63 (1971)].
[19]J. Marchunt and R. Gallibard, C.R. Acad. Sci. (Paris), Ser. B **281**, 261 (1975).
[20]J. P. Straley, J. Phys. C **9**, 783 (1976).