Dynamic Scaling near the Percolation Threshold in Thin Au Films

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We report on measurements of both the ac conductivity (σ) and the ac dielectric constant (ϵ) of thin Au films near their percolation threshold for frequencies between 100 Hz and 10 MHz. In the critical regime, $\sigma \simeq \omega^x$ and $\epsilon \simeq \omega^{-y}$. We obtain $x = 0.95 \pm 0.05$ and $y = 0.13 \pm 0.05$, in agreement with a general relation $x + y = 1$. These results are significantly different from predictions based on finite size scaling of the dc conductivity or diffusion on independent percolation clusters, providing evidence for the importance of electron-electron interaction effects.

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Second-order metal-insulator transitions are characterized by a vanishing of the dc conductivity, $\sigma(q=0,\omega=0)$, when the transition is approached from the metallic side, as well as the divergence of the real part of the dielectric function $\epsilon(q=0, \omega=0)$, when approaching the transition from the insulating side. By study of the critical behavior of σ and ϵ one can distinguish between the different mechanisms responsible for the metal-insulator transitions, e.g., between Anderson localization[1] and percolation.[1-3] For the latter one expects $\sigma(0,0) \sim \xi^{-\mu/\nu}$ for metal concentrations, p, above the percolation threshold p_c, and $\epsilon(0,0) \sim \xi^{s/\nu}$ for $p < p_c$, where $\xi \sim |p - p_c|^{-\nu}$ is the normalized percolation correlation length[4,5] measured in units of some microscopic, constant length, a (i.e., the actual correlation length is thus $a\xi$). The exponent s also describes the divergence of the conductivity of a metal-superconductor mixture for $p < p_c$.[2,6]

Another quantity of interest is the average polarizability of the finite clusters ($p < p_c$) which diverges as $\xi^{s'/\nu}$, where[3,4,7-9]

$$s' = 2\nu - \beta. \tag{1}$$

It has been pointed out[3] that s should not be identified with s'.[6] The divergence of ϵ contains two important contributions: (a) anomalous diffusion within each cluster and (b) intercluster polarization effects. The calculation of s' involves only (a) whereas previous derivations of the dielectric constant concentrated on (b). A unified theory which takes into account both effects is still lacking. Recently there have been extensive efforts to determine the values of s and s'.[4,7,10-17]

The above exponents describe macroscopic averages taken over samples which are larger than ξ. Near p_c, many physical quantities scale as a power law of the length scale L on which they are measured (for $L \ll \xi$). The naive scaling approach, employing the Einstein relation, replaces the length by a time scale,[3,18] thus obtaining $\sigma(0,\omega) \sim \omega^x$, $\epsilon(0,\omega) \sim \omega^{-y}$ (for $\omega \gg \omega_\xi \sim \xi^{-(2+\theta)}$, where ω_ξ^{-1} is the diffusion time for a distance ξ and $x = \mu/\nu(2+\theta)$, $y = s/\nu(2+\theta)$]. Using the relation[19-21] $\mu = s$ in two dimensions and substituting the current values of the critical exponents,[4,13,15] we find $x = y = 0.34$. This is in disagreement with a general scaling relation

$$y = 1 - x, \tag{2}$$

which follows directly by assuming that $\sigma(0,\omega)$ and $\epsilon(0,\omega)$ obey scaling forms which have a single characteristic time scale (e.g., $1/\omega_\xi$). Equation (2) also agrees with the expressions given in the scaling theory of Bergman and Imry.[2] Using $s = \mu$ and Eq. (2) yields $x = 0.5$, $y = 0.5$ ($d = 2$). The noninteracting cluster theory,[3] which uses s' rather than s, also agrees with Eq. (2) and yields $x \simeq 0.32$ and $y \simeq 0.68$. The dependence of transport quantities on the length and time scales in the critical regime near p_c has thus far only been tested by computer simulations.

In this Letter we report on the first systematic experimental study of both the ac conductivity[22] and the dielectric constant ($\propto$ capacitance) in the vicinity of the percolation threshold ($p > p_c$ and $p < p_c$). Our results yield values for x (0.95 ± 0.05) and y (0.13 ± 0.05) which are in good agreement with Eq. (2), although they are significantly different from theoretical predictions discussed above. We suggest that these data indicate that the incorporation into the theory of both dynamical intercluster effects (or more generally electron-electron interactions) and the delay time due to anomalous dif-

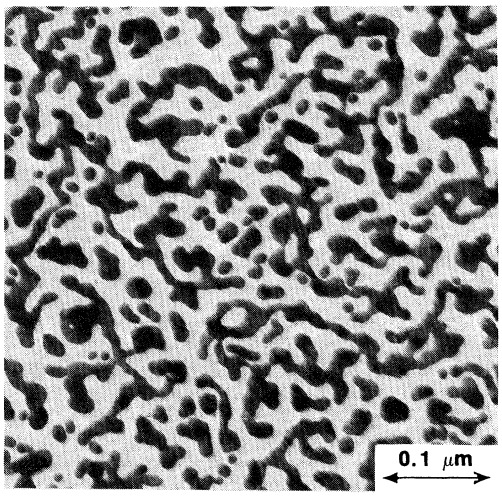

FIG. 1. Transmission-electron-microscope image of sample A-13, just on the insulating side with $a\xi \simeq 102$ nm. Typical cluster widths are about 10 nm.

fusion within the clusters is needed. In addition, our data verify for the first time that though the dielectric behavior of the conducting phase is very different from the insulator, there is no distinction between them at high frequencies in the critical regime, in agreement with theoretical considerations. We also find an explicit scaling function for $\sigma(0,\omega)$ on the metallic side, for both the high- and the low-frequency regimes.

The samples used in this study consisted of a series of Au films of varying thicknesses which were chosen to span the insulator-metal transition. The details of the fabrication process can be found in Laibowitz *et al.*[23] It is interesting to note that the nominal thickness difference between samples was generally less than 0.1 nm and a given fabrication run would consist of about 200 samples, a few millimeters on side. Such control of the thickness appeared to be important for close approach to the percolation threshold. In addition, the samples were studied by transmission electron microscopy (TEM) before electrical measurements were made. The TEM work was made possible by fabricating the samples on thin window substrates[24] and a typical micrograph is shown in Fig. 1. Such cluster pictures have also been digitized and using such data we have been able to derive the area dependence of the clusters,[25] identify the infinite cluster, and establish the correlation length from the micrographs, e.g., $a\xi$ for Fig. 1 is about 102 nm,[26] where a the microscopic channel width, is about 10 nm.

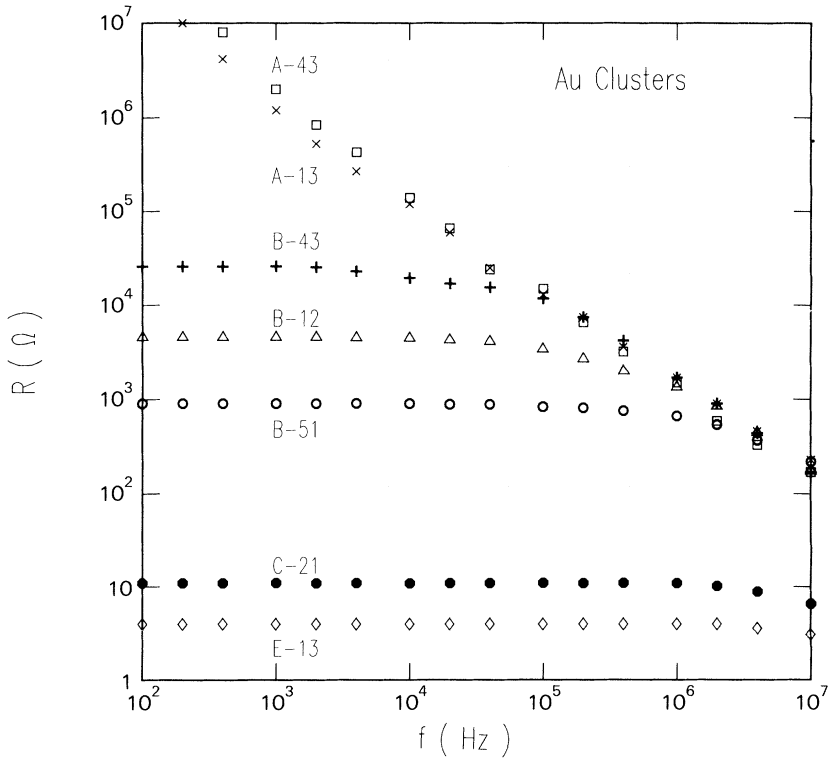

FIG. 2. Resistance data for both metallic and insulating samples. The curve labels refer to the specific position of the sample in the fabrication procedure, e.g., C-21 and E-13 are from the thicker (more metallic) sections.

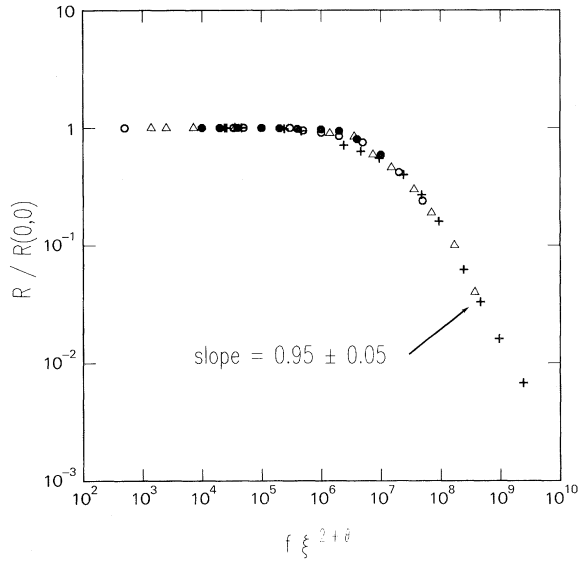

FIG. 3. The scaling function $g = R(0,\omega)/R(0,0)$ vs $f\xi^{2+\theta}$, for metallic samples from Fig. 2.

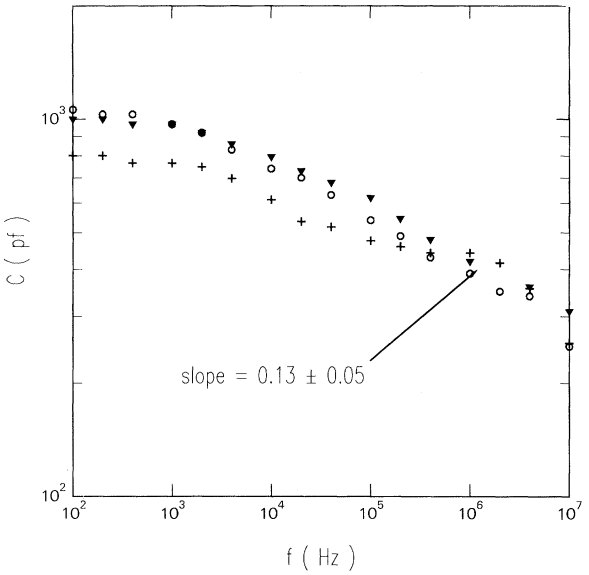

FIG. 4. Capacitance measurements just on the insulating side of the transition.

The room-temperature ac measurements were accomplished by use of standard techniques on samples in a four-terminal geometry. Efforts were made to use as low a measuring current as possible and heating effects were not observed in any of these studies. Insulating samples, i.e., $p < p_c$, with resistances in excess of 20 MΩ (~ 10 MΩ/□) were easily achievable indicating that in general tunneling and hopping contributions to the conductivity may be ignored in the more metallic samples. The results of the measurements of the sample resistance, R, versus applied frequency are shown in Fig. 2 for several samples. Notice that for the more homogeneous metallic samples (E-13 and C-21) the resistance remained almost constant throughout the entire frequency range. It should also be observed that the low-frequency behavior distinguishes between metallic and insulating (e.g., A-43) samples. The frequency dependence of the resistance $[R(0,\omega)]$ for the metallic samples is expected to be

$$R(0,\omega) = R(0,0)g'(\omega/\omega_\xi) = R(0,0)g(f\xi^{2+\theta}),$$

where $f = \omega/2\pi$. Figure 3 shows $g(f\xi^{2+\theta})$ for four metallic samples where for each sample ξ was chosen to give the best fit to the curve ($\theta = 0.86$; ξ for the most metallic sample, C-21, was chosen to be 1; for A-13, $\xi = 8$). These calculated values of ξ are found to be in good agreement with values found from analysis of the digitized micrographs.[25] In this work and in that of Kapitulnik and Deutscher[27] it has also been shown that the Au clusters may be described by percolation theory.

The resistance of the metal samples does not change significantly for $\omega < \omega_\xi$. For the insulating samples one expects $R \sim \omega^{-2}$ at low frequencies.[28] As shown in Fig. 2 the resistance of the insulating sample (A-43) clearly increases as the frequency is lowered but the asymptotic ω^{-2} behavior is not reached for frequencies above 400 Hz. At higher frequencies the electrons can scan a distance $L_\omega \sim \omega^{-1/(2+\theta)} < \xi$. On these scales the clusters are self-similar fractals. The average metallic density depends on L_ω and is larger the smaller is L_ω.[29] Thus the resistance will decrease with increasing frequency as shown in the data of Fig. 2 and can be described by the universal curve as shown in Fig. 3.[30] In fact, the value of x can be derived directly from the experimental data of Fig. 2.

Data for the capacitance (which is proportional to the real part of ϵ) are shown in Fig. 4. In general, these measurements are somewhat less accurate than those of the resistance and the relative error in y is large. Nevertheless using this data we have *independently* determined that $y \simeq 0.13 \pm 0.05$ which is significantly smaller than x, in agreement with Eq. (2) and thus in very good agreement with the resistance data.

In summary, our data appear to be in qualitative agreement with the dielectric behavior expected on both sides of the metal-insulator transition. It may be anticipated that when the electron-electron interactions become important, the screening length should increase, which implies a decrease of the

dielectric constant. This may help to explain why the exponent y that we obtain is smaller than that predicted by the single-particle theory.[3] However, a quantitative theory that takes into account the important dynamical and intercluster effects is still needed.

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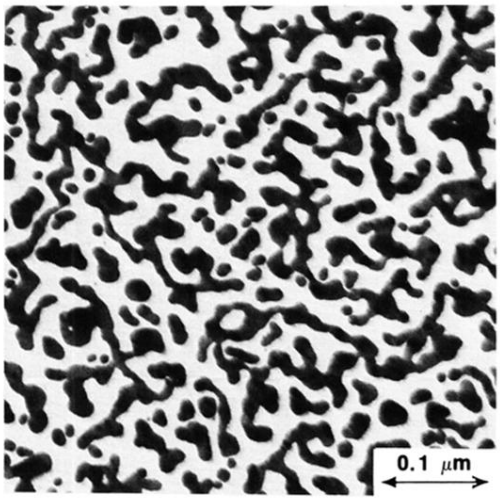

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