Conductivity and noise critical exponents in thin films near the metal-insulator percolation transition

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We report on experiments on thin silver films evaporated and ion milled at liquid-nitrogen temperature near the percolation threshold. We provide evidence for the universality of the critical exponent for the conductivity in two dimensions by showing that the critical exponents are the same in the two geometries studied. In contrast, the noise critical exponent κ is found to be different in the two model systems. For the case of the evaporated films $\kappa = 1.2 \pm^{0.3}_{0.7}$ in very good agreement with theory. For the ion-milled films, the critical exponent is $\kappa = 2.7 \pm 0.3$, which is much lower than expected from theory.

Recently, there has been considerable interest in the properties of thin metallic films near the metal-insulator percolation transition. This renewed attention arises from both the interest in determining under which conditions the lattice and continuum critical exponents for the conductivity can be nonuniversal[1] as well as determining critical exponents for the noise[2] and other nonlinear properties of these films.[3] In all these cases, the measurements represent a quantitative probe of the detailed structure of the percolating backbone and should allow us to examine in detail the conduction mechanisms present in these films near threshold. As shown by Halperin, Feng, and Seng,[1] the conductivity exponent t in two-dimensions is universal, and thus thin metallic films should have the same critical exponent as that of lattice percolation. In contrast, the noise exponent κ can be significantly different from system to system, since this critical exponent is quite sensitive to the small neck sizes which are present in the distribution to neck widths is continuum systems.[4] Experiments in two-dimensional model systems[5] have confirmed that at least some two-dimensional model systems are universal, exhibiting the same critical exponent as the lattice case. Experiments on the noise in thin metallic films are far less conclusive. Garfunkel and Weissman[6] have used sandblasted metal films of various materials to measure the noise exponent which they found to be in fairly good agreement with theory. In contrast, Koch, Laibowitz, Alessandrini, and Viggiano,[7] have measured a much lower critical exponent in ion-milled evaporated gold films at room temperature, a result that they ascribe to nonmetallic conducting paths in their films. In both of these experiments, the samples were of the Swiss-cheese type since a continuous metallic film was eroded mechanically until it reached the metal-insulator transition. One of the difficulties in these experiments is determining the critical threshold filling factor p_c; thus, rather than measuring both the normalized power spectrum S_v/V^2 and the resistance as a function of $p - p_c$, where p is the filling factor, it is much easier to relate the critical exponents t and κ directly through $S_v/V^2 \propto R^\omega$, where $\omega = \kappa/t$. This method thus assumes the universality of the critical exponent for the conductivity in two dimensions.

In this paper, we present experimental results on thin silver films at 77 K near the metal-insulator percolation transition. We show for realizations of both the Swiss-cheese and the inverse-Swiss-cheese models that the conductivity exponent is indeed universal in two dimensions and in good agreement with theoretical values. This is shown by determining the critical filling factor empirically by changing the assumed critical thickness near the percolation threshold, and looking at the quality of the fit. This procedure was found to yield consistent results which are remarkably close to the theoretical values for both the Swiss-cheese and inverse Swiss-cheese cases. This is to our knowledge the first determination of the universality of the conductivity exponent in a real system, rather than on a model system. Once the universality of the conductivity critical exponent was established, the normalized power spectrum was measured for both cases. We find that we can indeed unambiguously determine the difference between the Swiss-cheese-model and inverse-Swiss-cheese-model exponents for the noise. In the Swiss-cheese case, we find $\omega \sim 2.1 \pm 0.2$, in agreement with the results of Koch *et al.*[7] but lower than predicted in numerical simulations. In the inverse-Swiss-cheese case, we find $\omega \sim 0.9 \pm 0.2/0.1$, in reasonable agreement with the theoretical value of 0.87,[4] since this geometry is equivalent to the usual lattice case.

We made our resistance and noise measurements on evaporated silver films at 77 K in a cryogenically pumped thermal evaporator. The films were evaporated onto a glass substrate on a cryostat through a glass mask which generated a pattern of roughly 11 by 0.3 mm². Four-contact probes were located on the substrate to allow for the measurement *in situ* of both the resistance and the voltage noise. The films were evaporated at rates between 0.2 and 0.4 Å/sec. An ion-mill gun was located in direct line of sight below the sample in order to allow us to evaporate and to ion mill the sample and to measure the resistance and the noise as the films were being formed or etched, without breaking vacuum. Thus all our measurements were performed *in situ* on the same sample. We note that the ion-mill region was restricted to the center of the sample as our ion gun had a diameter of only 4 mm.

In practice, however, there is a significant amount of redeposition, during the milling process, of the silver inside of the glass mask which can create parallel channels to the sample near the percolation threshold. This can sometimes obscure the true percolation characteristics of the system near threshold. Thus, when measuring the resistance of the thin film as it was eroded, we simply evaporated a thick film, removed the mask, and then proceeded to ion mill it. This did not change the results, and eliminated the possibility of difficulties with parallel channels. We used slow milling rates (~ 10 Å/min) in order to control the film thickness close to threshold. Films were evaporated at low temperatures in order to limit the surface mobility in the film. Silver was chosen for its relative difficulty to form oxides as well as for the extensive characterization of silver films by the surface-enhanced Raman spectroscopy field.[8]

We begin by making the simplest possible model for our evaporated and milled films. In the first case, one can envision the nuclei that form the first monolayer of the films to be formed by circular disks of radius r and thickness d. Once these disks begin to overlap, a conducting backbone forms. This case is analogous to the inverse-Swiss-cheese model. For the ion-milled films, the geometry can be thought of as a metallic film in which holes are located at random as in the Swiss-cheese model. Thus, in this simple picture, our experiment covers the two most common models for continuum percolation, for which noise critical exponents have been explicitly predicted. Since we are unable to study the morphology of our films at 77 K we have to rely on room-temperature studies of the films. Transmission-electron-microscopy (TEM) analysis of our evaporated films near p_c showed that below p_c the films were circular droplets. When touching, these would form necks with a broad distribution of neck sizes. If compared with gold films,[7] our films tended to be less stringy and with more circular holes than gold films. Of course, these are only extrapolations that the morphology of the films did not change when warmed up. However, scanning-tunneling-microscopy studies of thicker silver films[8] do indicate that little structural change occurs from 77 to 300 K, although the films studied were thicker than those used in our experiments.

The $1/f$ noise was measured using a PAR-113 low-noise amplifier, the output of which was filtered and then fed into an HP3582A spectrum analyzer. The noise was measured between 0.2 and 20 Hz, with 64 averages performed for each resistance for which the noise was measured. The noise was always found to follow a $1/f^\alpha$ dependence with $\alpha \sim 1.1 \pm 0.06$. The normalized noise power spectrum was found to be independent of current. A calibrated quartz-crystal monitor was placed next to the sample to monitor the nominal thickness of the films during evaporation.

Figure 1 shows the resistance versus thickness plot for an evaporated film measured as it was slowly evaporated. While one might assign the origin of the observed divergence to percolating characteristics, one has to keep in mind that other conduction mechanism such as hopping or thermally activated tunneling can provide additional transport paths. In fact, one would expect these to be dominant before a metallic backbone forms. A second
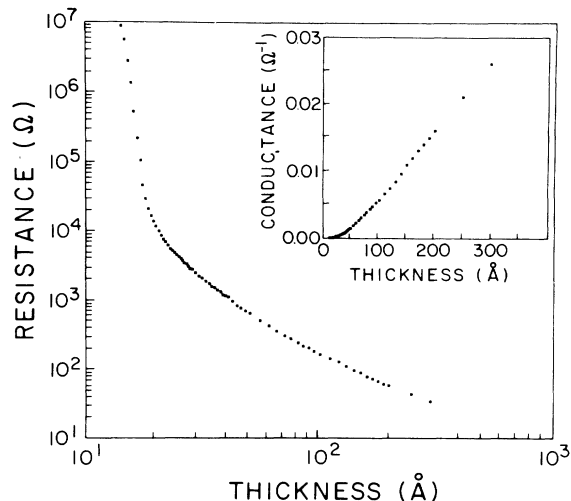

FIG. 1. Resistance vs thickness plot for a typical evaporated film. The inset shows the same data except that the conductance instead of the resistance is plotted.

difficulty with Fig. 1 is how to determine at which thickness the film no longer grows by forming a backbone of islands but rather grows uniformly. It has to be kept in mind that the nominal measured thickness is in fact incorrect since the crystal monitor assumes a constant area of coverage. Palevski et al.[9] have presented evidence that near the critical thickness d_c, the fraction of area coverage varies linearly with d, the average measured thickness which implies $(p - p_c) \propto (d - d_c)$ at least near p_c. We assume such a relationship is indeed valid and find our results consistent with such an assumption. This is in fact justified by the way in which the thickness monitor determines the thickness of the film by measuring the mass change on the substrate. If the film is uniform, the measured thickness will be $t_a = \Delta M / A\rho$, where M is the mass, ρ is the density, and A is the area of the substrate. If less than a monolayer is evaporated, the film will grow as clusters of average area a, and the total area of coverage will be Na, where N is the number of clusters. Then, the true thickness $d = \Delta M / Na\rho$ and $t_a = (Na/A)d = pd$. Then, the measured thickness is proportional to the area coverage of the film independent of the growth process for the clusters, and near p_c the film resistance should scale as $R \propto (p - p_c)^{-t} \propto (d - d_c)^{-t}$. Then, we just need to determine the true metallic threshold where the film first forms a connecting backbone.

In Fig. 2 we show the same data as in Fig. 1 plotted now versus $d - d_c$, where we have changed the assumed value of d_c for each of the three curves. As seen in Fig. 1, continuity is first observed for a value of film thickness of 14 Å. The three curves of Fig. 2 correspond to critical thickness values of $d_c = 15$, 16, and 17 Å, respectively. Two aspects are quite remarkable about this adjustment: First, despite the narrow range of variation of d_c, only the curve for $d_c = 16$ Å is close to a straight line; second, a least-squares fit to this curve yields an exponent of 1.35 ± 0.07, in remarkable agreement with the critical exponent for
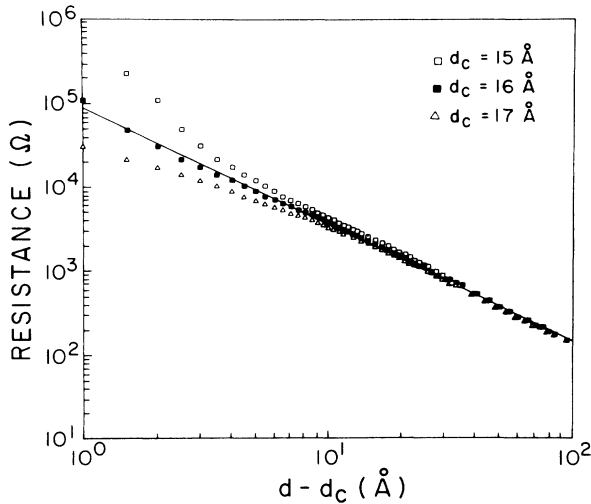
FIG. 2. Resistance vs $d - d_c$ plot for three different values of d_c as indicated in the figure for an evaporated film (realization of inverse-Swiss-cheese model). The solid line corresponds to a least-squares fit of the points for which $d - d_c < 20$ Å where cluster growth should still be horizontal.
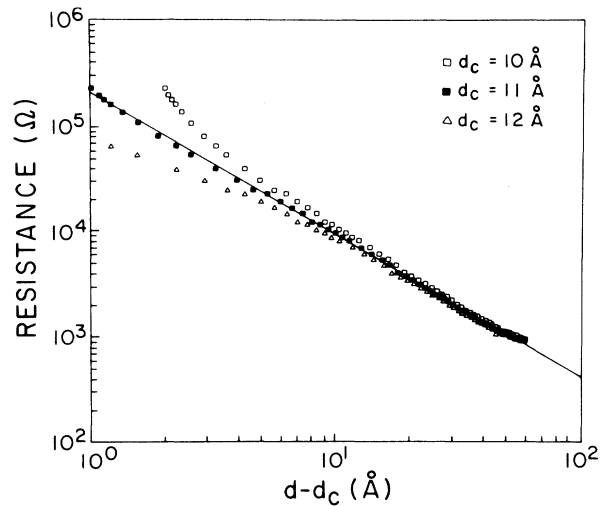

FIG. 3. Resistance vs $d - d_c$ plot for three different values of d_c for an ion-milled film (realization of Swiss-cheese model). The values of d were inferred from the known thickness and the ion-milling rate and time.

transport in two dimensions.[10] In fact, we have obtained similar data in over 20 separate evaporations which yield an average for the critical exponent t of 1.33 ± 0.04. This gives us confidence that we are indeed measuring the critical exponent for electric transport in our films. We note two additional features of our data. First, if the experiment is performed at room temperature, the data are not as reproducible, neither from run to run, nor in yielding any consistent data such as that of Fig. 2, an effect we attribute to diffusion of adatoms on the film surface. Second, since we could erode the sample and then evaporate again, we could check on the reproducibility of the results. After the first evaporation, subsequent evaporations yielded resistance values that differed by less than 1% once the differences in d_c between runs were subtracted. Typically, the first evaporation yielded a threshold for conduction between four to five angstroms above the value obtained in subsequent evaporations.

Exactly the same procedure yields consistent results when the same films are ion milled after evaporation of a sufficiently thick film. In this case, the thickness is determined from the ion-milling time, the film thickness, and assuming the same milling rate as for thick films. The resistance versus $d - d_c$ plot for the ion-milled case is shown in Fig. 3. Once again, the threshold value for metallic conduction has been varied 1 Å at a time for each of the three curves. As in the case of the evaporation, the curves are quite sensitive to the choice of d_c, and the best fit yields $t = 1.33 \pm 0.05$, which is remarkably close to the value expected for electric transport in two dimensions, and provides confirmation for its universality since the two cases are clearly of a different nature. We point out that in the great majority of the samples studied, we have found a value of t consistently higher than the theoretical accepted value, although this value was always within the

experimental error.

In Fig. 4 we show the measured noise as a function of resistance. Rather than showing the spectral density at a single frequency, we plot the average of the normalized spectral density times the frequency for the range of frequencies between 1 and 20 Hz where the noise was always of the form $1/f$ for all the resistance values to be considered. When the films were evaporated, the resistance would drift to a lower value quite slowly, which can be a
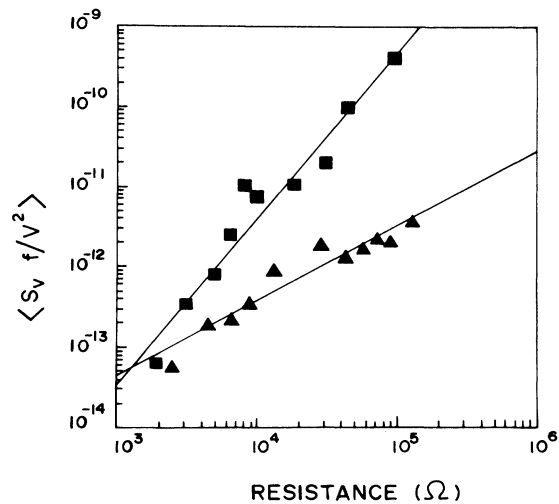

FIG. 4. Average of the normalized noise spectral density times the frequency between 1 and 20 Hz for both ion-milled (filled squares) and evaporated films (filled triangles) as a function of the resistance of the films.

spurious source of $1/f$ noise. Thus, we had to wait until this drift stopped before beginning a measurement. When the films were ion milled, the sample resistance relaxed rapidly to a lower value, when the ion milling was stopped, due presumably to heating effects. The large numbers of averages performed at each resistance and the low frequencies involved limited the number of points that could be measured for the same sample, and thus prevented us from doing a good fit of S_v/V^2 as a function of $d - d_c$ as in the previous figures. For the evaporated films we obtain $\omega = 0.9 \pm^{0.2}_{0.1}$, implying that $\kappa = 1.2 \pm^{0.3}_{0.1}$, in good agreement with the predictions of Tremblay, Feng, and Breton.[4] For the ion-milled films we obtain $\omega = 2.1 \pm 0.2$ or $\kappa = 2.7 \pm 0.3$. This value is in clear disagreement with simulations.[4]

The result for the inverse-Swiss-cheese-type case is unambiguously much lower than that for the Swiss-cheese-type case, despite the fact that the scatter is in good agreement with theoretical predictions. This difference between the two cases gives us further confidence in our conclusion about the universality of the conductivity exponent. The Swiss-cheese case is more difficult to interpret. Koch et al.[7] have obtained a very similar value for κ in their gold ion-milled films measured at room temperature. They attributed this difference to hopping as a parallel conduction mechanism not taken into account in simulations as well as variations in the distribution of neck widths. The coincidence of the two results would seem to rule out hopping since it would be quite surprising if two parallel noise mechanisms like metallic and hopping conduction with opposite temperature dependences would still yield a similar noise exponent at such widely different temperatures. It would also seem unlikely that both types

of films at different temperatures would have exactly the same distribution of both strengths. In fact, all that is required is that the distribution be fairly broad as observed in our films. We believe that a more fundamental difficulty remains unresolved. Recent numerical simulations yield values closer to our experimental results,[11] but it is not clear what the origin of the difference is. We also suspect that in order to properly calculate the noise, size effects have to be taken into account. In fact, we find it surprising that these have no effect on the conductivity exponent for at least one of the cases above, since the simplest arguments[12] would suggest that at least for the Swiss-cheese-type case they should be of importance.

In conclusion, we have presented experimental evidence for the universality of the conductivity critical exponent in two dimensions for both Swiss-cheese and inverse-Swiss-cheese models using percolating silver films at 77 K near the metal-insulator transition. The critical exponents for noise have been found to be quite different. In the case of the inverse-Swiss-cheese model good agreement is found between theory and experiments. For the Swiss-cheese case, the experimental critical exponent was found to be much smaller than in simulations. The similarity between our exponent and that of Koch et al.[7] suggests that important considerations are being left out of the calculation of the exponent for the noise.

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