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Resistivity-noise measurements in thin gold films near the percolation threshold

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Thin gold-film samples near the percolation threshold were fabricated with a resistance range from 10 to 10^8 Ω that had an equally large range of $1/f$ noise. The conduction mechanism and microscopic source of the noise changed from metallic to hopping as the sample resistance increased. Ion milling was used to increase the resistance of individual samples through the metal-insulator transition, and the measured $1/f$ noise, S_V/V^2, scaled as $R^{2\pm 0.1}$ on the metallic side.

$1/f$ noise can be a sensitive probe of the self-similar nature of a percolating metal film near the metal-insulator transition. If the resistance of a film fluctuates with time, each element of the film will contribute to the total noise magnitude proportional to the fourth power of the local current density through that element.[1] In this way the fractal properties of the percolation pattern, which are reflected in the local current density, can be probed. Fortunately, the resistance of a thin metallic film does fluctuate with time. The source of this noise ($1/f$ noise) is equilibrium thermal fluctuations that change the positions of the atoms and defects in the film, and hence change the local resistivity.[2]

Rammal, Tannous, Breton, and Tremblay[1] have introduced a new exponent κ that relates the magnitude of the normalized $1/f$ noise power to the filling factor on the metallic side of the metal-insulator transition,

$$S_V/V^2 \propto (p-p_c)^{-\kappa} \ , \tag{1}$$

where p is the filling factor and p_c is the critical filling factor. The resistance will also scale with the filling factor like

$$R \propto (p-p_c)^{-t} \ . \tag{2}$$

Both exponents depend on the fractal dimension of the percolating pattern. Testing either of these two relationships on experimental systems requires knowledge of the filling factor, which is difficult to obtain experimentally. Measuring the $1/f$ noise and the resistance simultaneously on the same sample can be easily done, and Eqs. (1) and (2) can be combined to give a unique prediction for the normalized noise as a function of resistance,

$$S_V/V^2 \propto R^{w} \ , \tag{3}$$

when the film is on the metallic side of the transition. Since in two dimensions the resistance exponent is often not particularly sensitive to the detailed nature of the percolating system, such a measurement basically measures the noise exponent κ.

Halperin, Feng, and Sen[3] have considered a class of random-void continuum percolation models in which the strengths of the bonds between the basic elements are continuously variable. This is unlike the lattice percolation model where the strengths of all bonds are identical. Conduction in two-dimensional thin metallic films seems well described by the continuum model, since the resistance between each pair of clusters in the film depends on the exact width of the metal neck connecting the two clusters. Unfortunately, in two dimensions the resistance exponent t is the same for both lattice and continuum models. The noise exponent w for a two-dimensional lattice percolation system is 1.0.[1] Garfunkel and Weissman[4] have predicted a value larger than 3 for w using a scaling argument and the random-void model. They have also measured the $1/f$ noise versus resistance of percolating films prepared using a sandblasting technique. Over a very small range in resistance (a factor of 3) they obtained a value of the noise exponent consistent with their model. Since then, Tremblay and Feng[5] have also predicted the resistance dependence of the $1/f$ noise for the random-void model and find a value near 4 for the noise exponent w.

To investigate these models and the actual self-similarity class of evaporated gold films near the percolation threshold,[6] we have measured $1/f$ noise versus sample resistance in these films as we changed the resistance in two ways. First, we have evaporated films of differing thickness, and second, we have increased the resistance of a single film by ion milling. We find the exponent w is 2.0 ± 0.1 when we used the ion-milling technique. When comparing different samples, the scatter in the measured noise spectra was too large to obtain a scaling relationship, although these results are similar to the ion-milling results. We have measured the temperature dependence of the sample resistance and noise to verify that the conduction mechanism and the microscopic source of the $1/f$ noise is metallic in the scaling region.

The samples were formed by vapor depositing thin gold films on a substrate surface between much thicker contact pads.[6] Si substrates with a Si_3N_4 surface layer were used, and the four contact pads were prepared by evaporating a 40-nm Au-Pd film through a photoresist lift-off mask. A lift-off mask was also used to pattern the sample size to typically 0.5 by 0.7 mm^2. Next the thin gold films were electron-beam evaporated at 0.02 nm/s onto a series of substrates until the average thickness was about 7 nm. The substrates were at room temperature during evaporation and had 200-nm-thick windows in them to allow TEM micrographs to be taken of the gold-cluster pattern (Fig. 1). In this thickness range, the gold migrates on the surface to form islands that are connected by narrow necks in a pattern that is self-similar and fractal on length scales larger then the basic cluster size.[7] About 50 samples were evaporated simultaneously in each run. The substrates were positioned such that the angle subtended by each substrate with respect to the evaporation source was different. The average gold thickness deposited was changed continuously by about 1 nm over the entire array of samples. This resulted in a

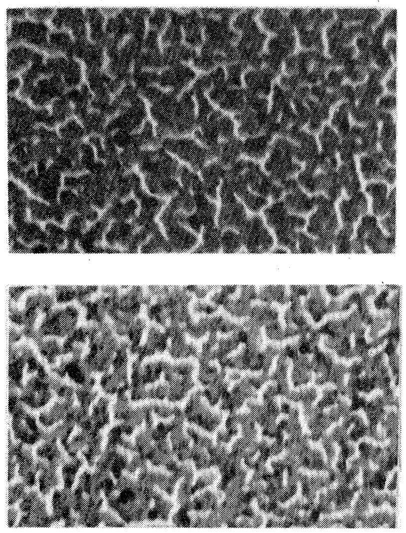

FIG. 1. Electron micrographs of a sample before (above) and after (below) ion milling. Shown area is 1.0×0.71 μm^2. The upper micrograph was made on a 300-Ω sample, and the lower micrograph was the same sample after ion milling. The final resistance was $> 10^7$ Ω.

resistance range from about 10^1 to 10^8 Ω for each run and satisfactorily covered the metal-insulator transition.

The $1/f$ noise was measured in 31 samples from several runs (Fig. 2) with two PAR 113 preamplifiers and an IBM PC that calculated the cross-power spectral density of the correlated noise between the two amplifier chains.[8] In samples with resistance less than about 10^5 Ω, the noise was measured by current biasing and measuring the voltage noise across the film. No significant differences were found using two- or four-terminal measurement techniques even for samples having resistances less than 10^2 Ω. In samples with resistances less than about 10^3 Ω, two PAR 1900 transformers were used to match the preamplifiers to samples. Samples having resistances greater than about 10^5 Ω were voltage biased and the current noise was measured using a resistor in series with the sample. In all cases the normalized $1/f$ noise power S_V/V^2 or S_I/I^2 was independent of the applied current or voltage bias. These measurements were all made at room temperature.

To assist in identifying the conduction and noise mechanisms, we measured the temperature dependence of the resistance and noise in 10 samples. Figure 3 plots the resistance and normalized $1/f$ noise for the two samples indicated with triangles in Fig. 2. High-resistance samples always decreased in resistance and noise as the temperature was increased, and lower-resistance samples generally displayed resistance and noise increases with increasing temperature.

To obtain higher-quality data near the metal-insulator transition without large sample-dependent variations, we placed single samples in an argon-discharge ion-milling machine where they were milled to increase the resistance in a controlled way. The gold was removed in a series of short ion-milling steps, each about 0.3 sec long, in order to control the decrease in the average film thickness. The amount of gold removed in each step was about 10 pm. The resistance and $1/f$ noise were measured *in situ* between each milling step. As more material was removed, the resistance increased more rapidly per milling operation, since the filling factor p was approaching the critical filling factor p_c. To compensate for this effect, the ion-accelerating potential (i.e., milling rate) was progressively reduced from an initial value of 350 to 70 V as the sample resistance increased. To prevent electrostatic charging of the film and destructive arcing during the milling operation, the argon-ion beam was neutralized with a tungsten filament, and all pads of the film were well grounded. This technique was used to reliably and predictably increase a

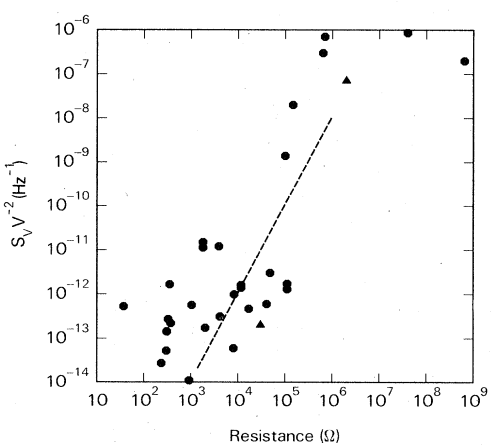

FIG. 2. Normalized $1/f$ noise S_V/V^2 (or S_I/I^2) vs sample resistance for all 31 samples measured. The temperature dependence of the noise is plotted in Fig. 3 for the two samples indicated as triangles. The dashed line is the result of the ion-milling experiment shown in Fig. 4.

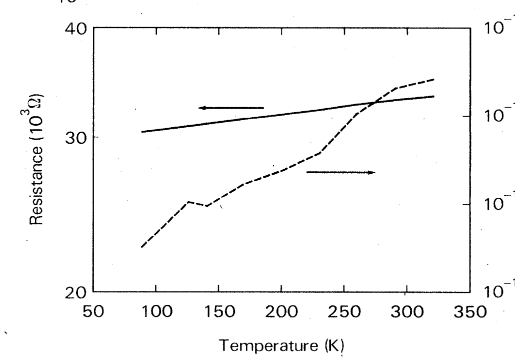

FIG. 3. Resistance and normalized $1/f$ noise S_V/V^2 (or S_I/I^2) for two samples vs temperature. The frequency was 10 Hz. The lower figure is for a metalliclike sample and the upper figure for an insulating sample.

film's resistance. Since the $1/f$ noise was large in this resistance range, the noise was measured using a two-terminal technique and one amplifier.

Figure 4 plots the normalized $1/f$ noise at 10 Hz, S_V/V^2, versus the measured sample resistance for one sample. Noise versus resistance measurements were made *in situ* on three samples. The value of normalized noise power S_V/V^2 for all three samples was about 10^{-12} Hz^{-1} at 10^4 Ω and 10 Hz. The normalized noise power on all samples varied as did the sample resistance R to the 2.0 ± 0.1 power. The dashed line on Fig. 4 indicates these results. The frequency exponent of the noise $d(\log S_V)/d(\log f)$ was -1.0 ± 0.1 and did not change during the ion-milling operations.

The inset of Fig. 4 is a histogram of the measured widths of the necks connecting larger clusters of gold. This was obtained from a micrograph similar to that shown in Fig. 1 that was subsequently ion milled and remeasured. The minimum neck width, about 4 nm, is larger than the resolution of the TEM and probably represents the minimum width a neck can be before surface tension causes the neck to open and form two unconnected clusters. The maximum neck width, about 16 nm, is characteristic of the maximum width of the gold clusters. The histogram of neck widths was systematically shifted by about 1 nm toward smaller neck sizes after the sample was ion milled. The distribution after ion milling was very similar to samples with the same resistance prepared without ion milling.

We now discuss the results. The overall $1/f$ noise versus resistance data plotted in Figs. 2 and 3 can be sorted into two types, metallic and insulating. The resistance at the critical filling factor p_c is about 10^5 Ω.[6,7] At values of R less than 10^5 Ω, the film is metallic, and the normalized noise was about 10^{-13}–10^{-11} Hz^{-1}. The conduction and noise mechanisms in this resistance range were characteristic of metallic conduction, since both increased with increasing temperature[2] almost identically to that measured for dirty gold films. This is the region where the geometrical theories of percolation apply. On the insulating side of the metal-insulator transition, or above 10^5 Ω, the noise is much larger than on the metallic side. The high-resistance samples had resistance and $1/f$ noise magnitudes that decreased exponentially with increasing temperature. This is characteristic of hopping conduction. The microscopic noise source is now related to electron trapping in the insulating regions between the metallic islands.[9] This type of source is not necessarily monotonic with temperature but generally decreases as the temperature is increased.[9] Two films with resistances near 1×10^5 Ω had temperature dependencies that did not monotonically increase or decrease with temperature, unlike the films mentioned above, which is strong evidence that the films were very near p_c.

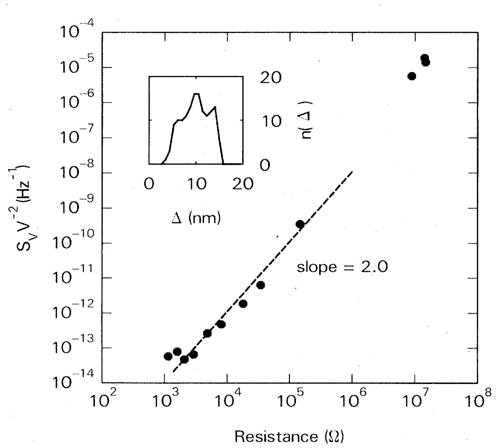

FIG. 4. Normalized $1/f$ noise S_V/V^2 vs resistance for a single sample measured between ion-milling operations. The frequency was 10 Hz. Inset: Histogram of neck widths in nm for 120 necks measured before ion milling.

There is significant scatter in the measured noise spectra for samples having almost identical values of resistance throughout the entire range of resistance, but particularly on the metallic side of the transition. The maximum correlation length ξ in these samples is about 100 nm, which is still 10^4 times smaller than the sample size,[7] so that finite size effects should not be important. The ion-milling experiment effectively eliminated the sample-to-sample variations in the $1/f$ noise. It allowed measuring the noise for many values of resistance in the range between 10^3 and 10^5 Ω, where the filling factor was close to the critical filling factor, and geometric scaling theories apply. The results of the ion-milling experiment when plotted against all the unmilled samples (Fig. 2, dashed line) strongly suggest that the intrinsic noise sources and magnitudes are not changed by the ion-milling operation.

The results of our measurements ($w = 2.0 \pm 0.1$) fit neither the lattice nor random-void models of percolation. This is not unexpected, since the bonds between the elements of a thin gold film are variable, unlike the lattice model, and the basic assumption of the random-void model, a uniform distribution of neck widths down to zero neck width, is also not satisfied. In our thin gold films there is a minimum neck width (Fig. 4, inset) which is about $\frac{1}{4}$ of the basic cluster size. Secondly, neither of these models include the possibility of hopping conduction or hopping noise, which is present in our gold samples. It is striking that the average noise versus resistance line in Fig. 4 could be extended into the insulating region with only a small change in slope.

We have measured the $1/f$ noise of evaporated gold films and have demonstrated that this system of microscopic islands connected by necks and hopping paths behaves neither as a lattice system nor as a random-void-type continuum percolating system. The temperature dependence of the conductivity and $1/f$ noise have been shown to be a sensitive tool for identifying the type of conduction present in a percolating film. The transition from metallic to thermally activated conduction near the metal-insulator transition has been shown to identify the transition. Finally, measurements of the conductivity versus filling factor near the metal-insulator transition will be affected by the nonmetallic conduction paths through the film. For instance, the changeover from metallic to hopping conduction across the film is a function of temperature when the filling factor is very near to the metal-insulator transition. Any complete theory of electron transport in microscopic percolating systems should include the possibility of thermally activated electron hopping.

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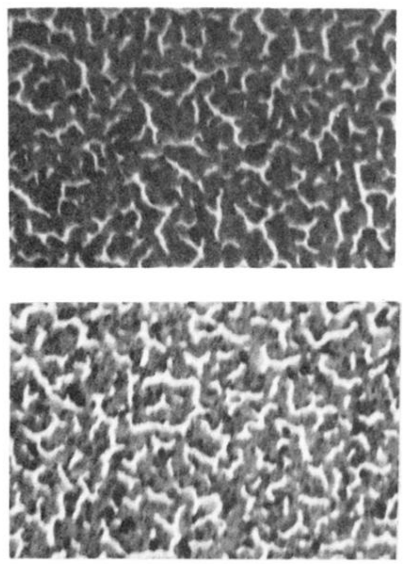

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