

Third-harmonic generation in semicontinuous metal films

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The weak nonlinear electrical response of two-dimensional semicontinuous Ag and Au films was measured over three decades of sheet resistance. The third-harmonic component generated by the films, due to local Joule heating, is interpreted as a measure of the fourth moment of the current distribution. Higher harmonics corresponding to higher moments of the current distribution were also observed. Under certain conditions, measurement of the third harmonic lends the same critical exponent as that of $1/f$ noise, which obeys a power-law dependence $S_R \propto (p - p_c)^{-\kappa}$, where S_R is the mean square of resistance fluctuations, p the surface coverage fraction, and p_c its critical value where the metal-insulator transition occurs. The film resistance obeys another power law $R \propto (p - p_c)^{-t}$, hence $S_R \propto R^w$ with $w = \kappa/t$. We have found that $w = 1.2 \pm 0.1$ for Ag samples, which is somewhat higher, but close to the lattice percolation value (random resistor network: $0.82 < w < 1.05$). For the Au samples, $w = 1.65 \pm 0.15$, which is higher than the lattice percolation exponent and lower than the Swiss-cheese-model continuum percolation estimate ($w > 4.5$). Comparison of our data on gold films with $1/f$ noise measurements on similar samples suggests the equivalence of these two techniques in probing the microgeometry details.

INTRODUCTION

The exact microgeometry of a percolative film can be probed by quantities whose critical behavior is nonuniversal. This is the case for $1/f$ noise which measures the fourth moment of the current distribution. Rammal *et al.*^{1,2} have introduced a new critical exponent κ that describes the divergence of the noise power $S_R \propto (p - p_c)^{-\kappa}$, where p is the filling factor and p_c the critical filling factor (at the metal-insulator transition). The resistance itself is described by another power law $R \propto (p - p_c)^{-t}$. Combining these two power laws yields $S_R \propto R^w$, where $w = \kappa/t$.

As in two-dimensional (2D) systems the resistance critical exponent t is practically universal, i.e., not sensitive to the exact microgeometry,³ the measurement of S_R provides the value of the noise exponent κ . The theoretical value for lattice percolation [random resistor network (RRN)] is $w = 0.86$ (Ref. 2), and the upper and lower bounds are 1.05 and 0.82, respectively.⁴ Different values are expected for continuum percolation. For the Swiss-cheese model w is in the range of 4.7 to 6.2.⁵ Bergman⁶ has calculated the sensitivity of this exponent to some details of the microgeometry, showing that experimental values should not be smaller than that of the lattice percolation. For ion-milled gold films Koch *et al.*⁷ obtained $w = 2.0 \pm 0.1$. Measurements of Al, In, and Cr films⁵ yielded w values in the range of 5 to 8. $1/f$ noise measurements are rather delicate as the noise power is inversely proportional to the sample volume and should be separated from other noise sources. This method is thus limited to small samples and noisy systems. An alternative measurement of the fourth moment of the current distribution, by Joule heating, was recently suggested by Dubson *et al.*⁸ A finite temperature coefficient

$\beta = (1/R)(dR/dT)$ results in the generation of higher harmonics.

We have applied the third-harmonic-generation technique on semicontinuous Ag and Au films using low-frequency electrical measurements. The third-harmonic coefficient is found to scale with the film resistance R as R^x , $x = 3.2 \pm 0.1$ and $x = 3.65 \pm 0.15$ for the Ag and Au films, respectively. Under certain conditions, which are discussed below, x may be interpreted as the critical exponent of the fourth moment of the current distribution, i.e., $x = 2 + w$. Assuming that this is the case in our films, we find that $w = 1.2 \pm 0.1$ and $w = 1.65 \pm 0.15$ for the Ag and Au films, respectively. The measured value of w for the Au films is in fair agreement with previous $1/f$ noise measurements.⁷ For the Ag films, w is higher than, but close to the upper bound of the RRN model. The harmonic-generation method allows a measurement of the fourth moment down to relatively low sheet-resistance films, for which the $1/f$ noise would be very small, leading to a more accurate determination of the exponent w in a clearly metallic regime. Another advantage of this method is that it is more general: while the $1/f$ noise measurement leads only the fourth moment of the current distribution, higher moments can be obtained through measurements of higher harmonics, which we have indeed detected. However, if the Joule heat is spread on relatively large areas, the third (and higher) harmonics are not directly connected to the corresponding moments of the current distribution, and the interpretation of the data is much more complicated.

The film resistance probes the second moment of the current distribution as can be easily seen from conservation of energy: $I^2 R = \sum i_\alpha^2 r_\alpha$, where R is the film resistance, I the total current, and i_α and r_α are the local current and resistance, respectively. The relative resis-

tance noise $S_R = \langle \delta R^2 \rangle / \langle R \rangle^2$ probes the fourth current distribution moment^{1,2} and is given by $\Sigma i_\alpha^4 \langle \delta r_\alpha^2 \rangle / (\Sigma i_\alpha^2 r_\alpha)^2$, where δr_α is the resistance fluctuations of r_α (assuming that δr_α and δr_β are not correlated: $\langle \delta r_\alpha \delta r_\beta \rangle = \delta_{\alpha,\beta}$). If all the elementary resistors r_α are identical and have the same value r , the above relation reduces to $\delta r^2 \Sigma i_\alpha^4 / r^2 (\Sigma i_\alpha^2)^2$. The relation between resistance fluctuations and thermal response was first recognized by Weissman and Dollinger.⁹ The local resistance change, assuming a linear thermal response, is

$$\delta r = r \beta \Delta T = \beta i^2 r^2 h(\omega, T), \quad (1)$$

where $h(\omega, T)$ defines the coupling between the local dissipation and the local temperature change (including the heat conduction, the specific heat, and the coolant and substrate influence). For an ac current $I = I_0 \cos(\omega t)$, the local resistance r_α can be written as

$$r_\alpha = r_0 + \delta r_\alpha \cos(2\omega t + \phi), \quad (2)$$

where the correction of δr_α to the first term was ignored as $\delta r_\alpha \ll r_0$, and ϕ is the phase shift between the heat production and the local temperature. The voltage across the sample is given by

$$V = IR = I_0 R_0 \cos(\omega t) + \frac{1}{2} I_0 \Delta R \cos(3\omega t + \phi). \quad (3)$$

This is the source of the third harmonic generated by the sample. Substituting δr and using $I^2 \Delta R = \Sigma i_\alpha^2 \delta r_\alpha$ yields

$$V_{3f} \propto I_0 \Delta R \propto \frac{1}{I_0} \Sigma i_\alpha^4 r_\alpha^2 \quad (4)$$

or

$$\frac{V_{3f}}{I_0^3 R^2} \propto \frac{\Sigma i_\alpha^4 r_\alpha^2}{I_0^4 R^2} \propto \frac{\Sigma i_\alpha^4 r_\alpha^2}{(\Sigma i_\alpha^2 r_\alpha)^2}, \quad (5)$$

where we have assumed that $h(\omega, T)$ is position independent, i.e., does not depend on α . Assuming again $r_\alpha = r$, one finds

$$S_R \propto \frac{V_{3f}}{I_0^3 R^2} \quad (6)$$

and V_{3f} should scale as

$$\frac{V_{3f}}{I_0^3} \propto R^{2+w}. \quad (7)$$

A more detailed calculation of δr in (1) yields the production of higher-order odd harmonics. If the zero current resistance is r_0 then (1) can be written in the form

$$\delta r = r_0 \beta h(\omega, T) i^2 (r_0 + \delta r). \quad (8)$$

Expanding δr in a power series yields

$$\begin{aligned} \delta r &= \frac{\beta h(\omega, T) r_0^2 i^2}{1 - \beta h(\omega, T) r_0 i^2} = r_0 \Sigma [\beta h(\omega, T) r_0] i^{2n} \\ &= \delta r_1 + \delta r_2 + \delta r_3 + \dots, \end{aligned} \quad (9)$$

where

$$\delta r_1 = r_0 [\beta h(\omega, T) r_0] i^2, \quad (10a)$$

$$\delta r_2 = r_0 [\beta h(\omega, T) r_0]^2 i^4, \quad (10b)$$

$$\delta r_3 = r_0 [\beta h(\omega, T) r_0]^3 i^6. \quad (10c)$$

Using again $I^2 \Delta R = \Sigma i_\alpha^2 \delta r_\alpha$ and $\Delta V = (1/I) \Sigma i_\alpha^2 \delta r_\alpha$, one finds that the third harmonic is generated by δr_1 , the fifth by δr_2 , the seventh by δr_3 , and so on. The $(2n+1)$ harmonic is thus given by

$$\begin{aligned} V_{(2n+1)f} &= (1/I) \left(\frac{1}{4}\right)^n r_0 [\beta h(\omega, T) r_0]^n \Sigma i^{2n+2} \\ &= I^{2n+1} \left(\frac{1}{4}\right)^n r_0 [\beta h(\omega, T) r_0]^n N^{-2n} (R/R_0)^{\alpha_n}, \end{aligned} \quad (11)$$

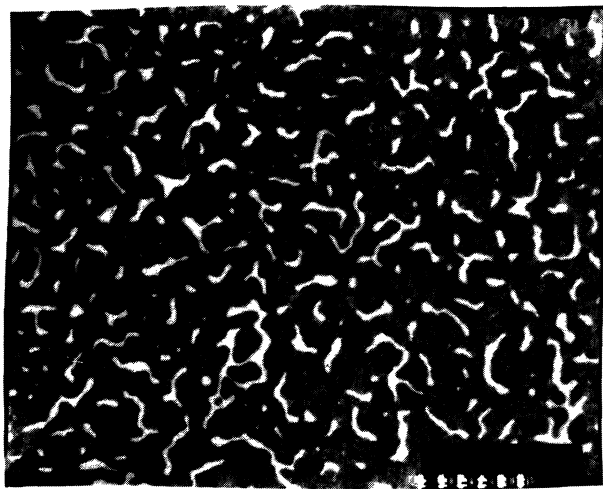
where N^2 is the number of resistors in the RRN model, R_0 is the sheet resistance of a continuous film ($p=1$), and α_n is the critical exponent of the $(2n+2)$ moment of the current distribution.

In the above derivation we have assumed that the temperature increase of each resistor r_α depends solely on i_α , ignoring the heat flow between adjacent resistors. Typically, a link which carries high current density causes the temperature increase of its surrounding links. The contribution of these links to the total resistance change $\Delta R = \Sigma i_\alpha^2 \delta r_\alpha$ depends on their number and their current densities. If ΔR is dominated by the high-current-densities links, Eq. (11) is valid and the harmonic-generation technique provides a direct measurement of the corresponding moments of the current distribution. In the opposite case (ΔR is dominated by the heated low-current-densities links), the harmonic-generation method measures some kind of the autocorrelation function of the second moment of the current distribution, where the exact form depends on the temperature profile. A fuller analysis requires the solution of the heat flow in such a system, which was not done yet as far as we know.

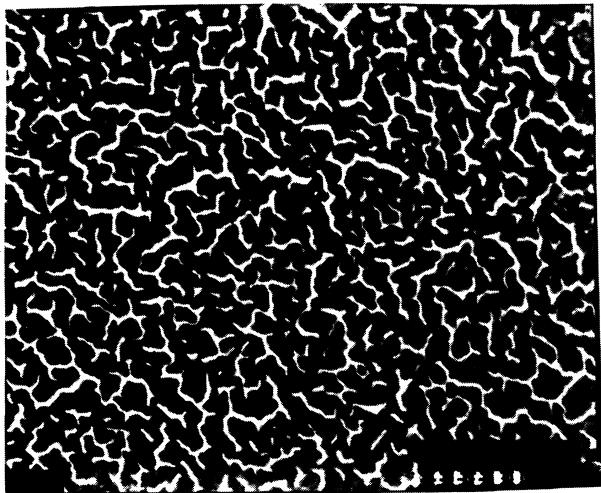
EXPERIMENT

Thin semicontinuous Ag and Au percolating films were evaporated under vacuum of 10^{-6} torr at a rate of 0.1 nm/s onto room-temperature glass substrates. Several samples with different surface coverage values were prepared simultaneously in each run. Their size was 4×1 mm². The samples were measured at room temperature, liquid-nitrogen (77 K), and liquid-helium (4.2 K) temperatures. The third harmonic generated by the samples was measured by an HP35660A two-channel dynamic signal analyzer, where channel *A* was directly connected to a resistive balance bridge and channel *B* to a series resistor measuring the ac current. The HP35660A internal source provided a monochromatic ac signal with maximum ratings of 3 V rms and 30 mA. The maximum voltage applied on the sample was thus 1.5 V rms, due to the bridge operation. Detection of the third harmonic was normally measured at source voltages of 0.1–3 V rms. This method provided a sensitivity of better than –120 dB, which was sufficient to measure the third harmonic of an almost continuous metal film. Most of the data were obtained at a frequency of 1.5 Hz and at room

temperature. Measurements at other temperatures (77 and 4.2 K) and frequencies (within the range 0.1–35 Hz) were performed in order to verify the independence of the critical exponent on those parameters. Resistance versus temperature measurements showed a positive temperature coefficient (metallic conductivity). The I - V characteristics of several samples at various temperatures show a linear dependence (Ohmic behavior) at low enough currents (the actual values are strongly sample dependent) and a weak resistance increase at higher currents (for example, above 1 mA for a 1-K Ω film), due to Joule heating of the entire sample. TEM micrographs of silver [Fig. 1(a)] and gold [Fig. 1(b)] films show percolative two-dimensional structures with an average channel



(a)



(b)

1000 Å

FIG. 1. TEM micrographs of semicontinuous metal films. (a) 7.5-nm-thick Ag film. (b) 6.5-nm-thick Au film.

width of 30 and 20 nm, respectively. Qualitative examination of the microgeometry shows the existence of some much narrower channels. They exist both in the Ag and Au films, with no clear qualitative difference between them [apart from the different surface coverage parameter, i.e., $(p-p_c)$ in the Ag sample is larger than that of the Au sample]. Identifying differences between the two microgeometries by investigating the TEM micrographs would be a complicated task requiring sophisticated image analysis. Moreover, differences in the exact microgeometry may result from the existence of photoresist or carbon film on the substrate, thus the TEM micrographs cannot always represent the precise structure.

RESULTS

We assume in our analysis that Eq. (11) is valid. Our main empirical reason for believing so is the fact that the average temperature rise of the film at a typical power level that we use is orders of magnitude smaller than the local temperature rise. The average temperature rise can be obtained from the change in the measured macroscopic resistance of the film. This is typically less than 0.1 K (less than 0.01% change in the macroscopic resistance; see Fig. 3 for $I < 1$ mA). On the other hand we show below that the observation of third-harmonic generation in liquid-helium-cooled Au films can only be explained by local temperature rise of at least 10 K. This large difference between average and local heating can only be understood if the fraction of significantly heated links is very small. Possible theoretical verifications for the validity of this assumption are discussed below.

The normalized third-harmonic amplitude ($B = V_{3f}/I^3$) of the silver films versus the room-temperature resistance R is shown in Fig. 2. The straight line yields a power-law dependence $B \propto R^{2+w}$, giving a critical exponent $w = \kappa/t = 1.2 \pm 0.1$. Substituting $t = 1.3$ yields $\kappa = 1.55 \pm 0.15$. The measured value of w is higher than, but close to the lattice percolation upper bound⁴ $0.82 < w < 1.05$. The third-harmonic amplitude at 77 K is almost twice that at room temperature and the critical exponent is unchanged. Cooling to 77 K was done by either direct contact with liquid nitrogen or with exchange

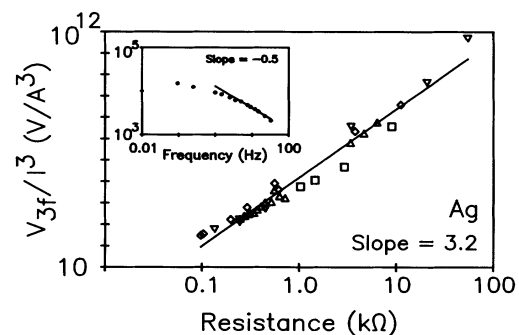


FIG. 2. The scaling of B for Ag films, $2+w = 3.2 \pm 0.1$ (logarithmic scales). The different symbols represent separate fabrication runs with several samples fabricated in each run. The inset shows the frequency dependence of B for one of the Ag films. The straight line represents $V_{3f} \propto \omega^{-1/2}$ (see Ref. 8).

gas; in both cases the third-harmonic amplitude was the same. This indicates that the heat flow from the weak links is primarily to the metallic clusters and that the heated areas are very small indeed; thus a hot-spot picture is appropriate. At 4.2 K no third-harmonic component was found using the same current range (the maximum current used in our measurements is reached by applying 1.5 V rms on the sample). The resistance-temperature coefficient was measured and found to become very small ($\beta < 10^{-5} \text{ K}^{-1}$) at these temperatures, thus local heating cannot change significantly the local resistance and a nonlinear response is not expected: as shown above, the third-harmonic amplitude is proportional to the resistance-temperature coefficient β and is thus suppressed. Above 30 K a linear temperature dependence is found up to room temperature with a constant slope $\beta = (1/R)dR/dT = 6.4 \times 10^{-4} \text{ K}^{-1}$, where R is the resistance at room temperature. The frequency dependence of B was also checked and is shown in the inset of Fig. 2, where again w is unchanged. This dependence is dominated by the heat-transfer mechanism and was discussed in Ref. 8. A power-law dependence $V_{3f} \propto \omega^{-1/2}$, resulting from a one-dimensional heat-flow picture (from the film into the substrate), is found only for a limited frequency range. This is another indication that the heat flow through the metallic channels is not negligible. An I - V characteristic of one of the Ag samples is shown in Fig. 3. The sample resistance increases with the current, showing that the nonlinear response is due to Joule heating. The resistance change is noticed only at higher currents than those used in the ac measurements (less than 1 mA in this case), showing that the heating in the latter case is really a local effect. Fitting the resistance change with a quadratic correction $V/I = R + BI^2$ yields $B = 10^5 \text{ V/A}^3$ while for the third-harmonic generation $B = 5.2 \times 10^4 \text{ V/A}^3$ at 1.5 Hz. Although the observed frequency dependence of B does not permit accurate extrapolation to $\omega = 0$ (see the inset of Fig. 2), it seems reasonable that the dc result would be regained in that limit. Thus in the third-harmonic method one also measures the onset of the nonlinear response.

The scaling of V_{3f} as a function of R for gold films is shown in Fig. 4. In this case the data are more noisy, but

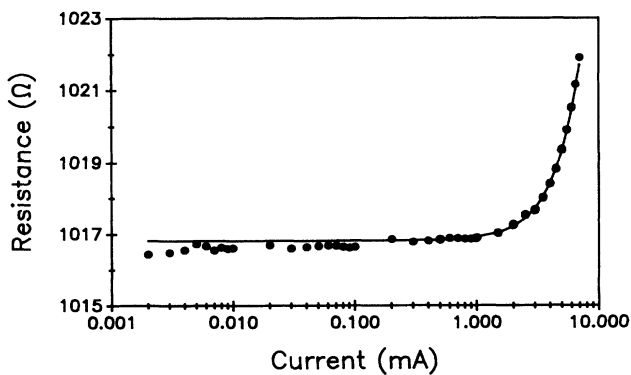


FIG. 3. I - V characteristic of one of the Ag films (semilogarithmic scale). A deviation from Ohmic behavior is observed at high currents due to Joule heating. The solid line represents the fit $V/I = R + BI^2$, $B = 10^5 \text{ V/A}^3$.

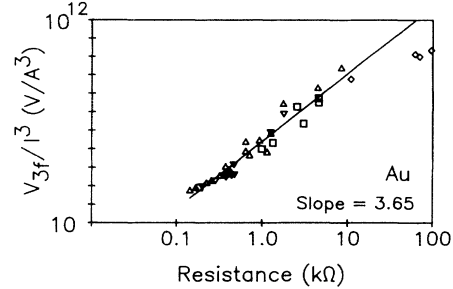


FIG. 4. The scaling of B for Au films, $2+w = 3.65 \pm 0.15$ (logarithmic scale). The different symbols represent separate fabrication runs with several samples fabricated in each run.

a power law with critical exponent $w = \kappa/t = 1.65 \pm 0.15$ is well defined up to $R = 10 \text{ K } \Omega$. In particular, the value of w in this case is definitely larger than that of the silver films. Substituting $t = 1.3$ yields $\kappa = 2.15 \pm 0.2$, in close agreement to the value obtained by Koch *et al.*⁷ using $1/f$ noise measurements on gold films. The microgeometry of Au films is thus different from that of Ag, and is not well described either by lattice percolation or by the Swiss-cheese model. Again no change in w was found for different temperatures and frequencies. The frequency dependence and the I - V characteristic of the Au films are similar to those of the Ag films. The magnitude of the third-harmonic component was enhanced at 77 K, like the case of the silver films. At 4.2 K third-harmonic generation was observed and the amplitude was smaller than the amplitude measured at room temperature (unlike the silver films where third-harmonic generation was suppressed at 4.2 K). The resistance-temperature coefficient of Au was measured and found to become very small ($\beta < 10^{-5} \text{ K}^{-1}$) below 15 K. The observation of a third harmonic at 4.2 K indicates that the temperature rise at the weak link is more than 10 K. Above 15 K a linear temperature dependence is found with a constant slope $\beta = (1/R)dR/dT = 6.7 \times 10^{-4} \text{ K}^{-1}$, where R is the resistance at room temperature. For a 200- Ω sample, this behavior was measured up to 400 K (the highest temperature reached). On the contrary, a 100-K Ω sample showed this behavior up to 320 K only; beyond that the resistance-temperature coefficient changed sign and became strongly temperature dependent. At 380 K, $\beta = -8 \times 10^{-3} \text{ K}^{-1}$, i.e., a negative coefficient whose absolute value is an order of magnitude larger than the metallic value. The third-harmonic coefficient of samples with resistances higher than 10 K Ω seems to saturate (unlike the case of the silver films). This saturation and the anomalous resistance-temperature coefficient of high-resistance films indicates that their conductivity is not purely metallic. We interpret this behavior in the following way: when a high current is applied to a weak link, the local temperature change is sufficient to excite local hopping, reducing the current density in the weak link and resulting in a weaker third-harmonic component. Such a hopping mechanism could be expected in a continuum percolation structure. For illustration, consider two metallic disks just touching each other. The point contact is a metallic weak link, while on both sides

the separation between the two metallic disks is extremely narrow, and hopping will be excited at relatively low temperatures.

Generation of higher odd harmonics measures higher moments of the current distribution, as discussed above. We have detected fifth and seventh harmonics on several high-resistance films, and verified that the normalized quantities V_{5f}/I^5 and V_{7f}/I^7 are constant, i.e., current independent. As discussed by Bergman,⁶ these higher moments are even more sensitive to the exact microgeometry and may be useful if such information is needed. To observe these harmonics on more metallic films, one would have to use a high-pass filter or a Fourier-transform analyzer with a much larger dynamics range, as the third-harmonic amplitude is much larger than the higher harmonics. From the ratios between the third, fifth, and seventh harmonics of one of the films, one may estimate the critical exponents of these higher moments:

$$\frac{V_{(2n+1)f}/I^{2n+1}}{V_{(2n-1)f}/I^{2n-1}} = \frac{1}{4}[\beta h(\omega, T)r_0]N^{-2}(R/R_0)^{\delta\alpha_n}, \quad (12)$$

where $\delta\alpha_n = \alpha_n - \alpha_{n-1}$. The prefactor $[\beta h(\omega, T)r_0]N^{-2}$ can be estimated from the regression of B versus R : $B = B_0R^{2+w}$, where for Au films we obtain $2+w = 3.65$ and $B_0 = 3 \times 10^{-6} \text{ V/A}^3$. The basic sheet resistance R_0 and the basic link resistance r_0 are both of the order of the sheet resistance of a continuous film, i.e., in the range 1–10 Ω for a 10-nm-thick film. The resistance-temperature coefficient β was measured: $\beta = 6.7 \times 10^{-4} \text{ K}^{-1}$. The value of N can be estimated by the ratio between the film size and the separation between two adjacent links, thus $N \approx 1 \text{ mm}/30 \text{ nm} \approx 3.5 \times 10^4$. Using the above values, the quantity $h(\omega, T)$ of a hot spot can be estimated: $h(\omega, T) \approx 10^9 \text{ K/W}$. We do not attempt to solve the heat-transfer problem in this case to calculate $h(\omega, T)$; however, by estimating the heat transfer to the glass substrate and the heat flow through the percolating metal film we found that $h(\omega, T)$ is of the above order of magnitude, and that the typical hot-spot size is of the order of 100–1000 nm (see the Appendix). A uniform sample heating would yield $h(\omega, T)$ smaller by several orders of magnitude, as can be seen in the dc measurements. We take this estimate as an indication for the existence of hot spots in the ac measurements. The relatively large size of the hot spots permits the assumption that $h(\omega, T)$ is position independent.

Close to the percolation threshold the infinite cluster is rather ramified, hence most of the mass of a hot spot belongs to dangling bonds. The existence of a high current density in one link suggests that it is connected to the infinite cluster by rather strong links, i.e., with low current densities. These arguments, together with the large value of $h(\omega, T)$ which indicates that the hot spots cover a small portion of the film, suggest that the validity of Eq. (11) is at least reasonable. The third harmonic may thus be interpreted as a measure of the fourth moment of the current distribution, and the critical exponent as $2+w$.

A measurement of the fifth and seventh harmonics of a 7-K Ω -resistance Au film is shown in Fig. 5. The ratio

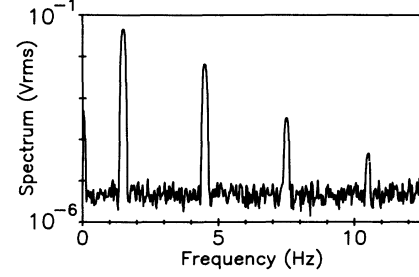


FIG. 5. Fourier spectrum of a 7-K Ω -resistance Au film, showing the odd harmonics generated by the film. The input signal is at 1.5 Hz and the corresponding Fourier component is the residual signal after the bridge. The applied current is 250 μA .

between the normalized harmonics (averaged for several input currents) is $(V_{7f}/I^7)/(V_{5f}/I^5) \approx 5 \times 10^6$ and $(V_{5f}/I^5)/(V_{3f}/I^3) \approx 5 \times 10^6$. Substituting the above values in the prefactor [see Eq. (12)] one finds that the difference between the critical exponents α_n is $\delta\alpha_n = 3.2$ for $R_0 = 1 \Omega$ and 3.5 for $R_0 = 10 \Omega$, for $n = 2$ and $n = 3$. A factor of 2 in the prefactor corresponds to a change of 0.1 in $\delta\alpha_n$, thus we conclude that $\delta\alpha_{2,3}$ is in the range $3.7 > \delta\alpha_{2,3} > 3.0$. The step between the second and third moments of the current distribution is $\delta\alpha_1 = 2.65 \pm 0.15$. These values are higher than the expected steps for lattice percolation,^{1,2,10} which are of the order of 1.9 for $\delta\alpha_1$ and 2 for $\delta\alpha_2$ and $\delta\alpha_3$. However, it is in agreement with the prediction that $\delta\alpha_1$ is smaller than $\delta\alpha_2$ and $\delta\alpha_3$. These estimates of the critical exponents show that for continuum percolation the moments of the current distribution diverge much faster than that of lattice percolation, as discussed by Bergman.⁶ Measurements of high moments of the current distribution as well as computer simulations are needed for further study of the critical exponents and their dependence on the microgeometry.

DISCUSSION

A comparison of our results on gold films with the $1/f$ noise measurements obtained by Koch *et al.*⁷ shows that in the metallic regime, where hopping is unimportant, these two methods yield the same critical exponent, w . If one excludes the last point in the $1/f$ noise measurements of Ref. 7 ($R > 10^5 \Omega$, i.e., the sample is very close to the percolation threshold), the resulting exponent is $w = 1.6 \pm 0.2$ (as we have estimated from the published data), in excellent agreement with our result. For films with sheet resistances higher than $10^5 \Omega$, hopping seems to be more important and the $1/f$ noise is larger. On the contrary, the third-harmonic signal is weaker in this regime, as discussed above. These opposite trends can be very useful in identifying the crossover between the purely metallic conductance and the hopping and tunneling regime. The comparison with the $1/f$ noise measurements also shows the advantages of the third-harmonic method. While using a simpler experimental setup we could measure samples with much lower sheet resistances, and the scattering of different samples is much

lower (in the $1/f$ noise measurements of Ref. 7 the critical exponent was measured on an ion-milled film; the scattering of the data of different samples could not provide a reliable value).

In summary, we have applied the new thermal-response technique based on third-harmonic detection on real materials and verified that local Joule heating is the important mechanism. The results are interpreted under the assumption that this is a direct measure of the fourth moment of the current distribution. A fuller theory, including the effect of the heat spread, is still lacking. Higher harmonics were also observed, thus higher moments of the current distribution are accessible using the same experimental setup. From the ratio between the third, fifth, and seventh harmonic of one of the Au samples we could estimate the critical exponent $3.7 > \delta\alpha_{2,3} > 3.0$, which is higher than the lattice-percolation value, and can be understood as due to the continuum nature of the Au films. We found out that the critical exponent depends only on the microgeometry and not on temperature nor on frequency. For Au films we obtained $w = 1.65 \pm 0.15$ in agreement with Koch *et al.*,⁷ measuring $1/f$ noise on similar samples. This agreement verifies the validity of this technique to measure the fourth current distribution moment. It also provides an overall verification for the physical assumptions used in the derivation of both the thermal response and the $1/f$ noise expressions. The measured value of w shows that Au films should be characterized by a continuum percolation description, but different from the Swiss-cheese model. A more flexible model, allowing cutoff in the minimum channel width, is required to describe the microgeometry of real materials. For Ag films, we found $w = 1.2 \pm 0.1$ which is closer to the lattice-percolation picture ($0.82 < w < 1.05$), indicating that the enhancement of w due to the continuum nature is less important. Silver films are thus preferable in measurements connected to lattice percolation and should be a better model system for those purposes than the commonly used gold films.

ACKNOWLEDGMENTS

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APPENDIX

The issue of the heat flow from a hot spot is beyond the scope of this work. However, from some general arguments one may estimate $h(\omega, T)$, the coupling coefficient between the applied power and the temperature rise at a hot spot. Two main contributions should be taken into account: heat flow from the surfaces of the film, and heat

flow through the metallic channels. The size of the hot spot depends on the efficiency of these two mechanisms, and may thus be estimated by the condition that these two contributions are equal.

The heat flow to the substrate of a hot spot of linear size L , assuming only one interface (i.e., in vacuum), is given by

$$J_s = \alpha L^2 (T_{av} - T_s), \quad (A1)$$

where α is the surface thermal conductance (given in units of $W/cm^2 K$), T_{av} the average temperature of the hot spot, and T_s the substrate temperature (which is assumed to be constant over the area of the hot spot).

The heat flow along a metallic channel of cross section A is given by

$$J_m = \frac{\kappa A}{L_m} (T_{max} - T_s), \quad (A2)$$

where κ is the thermal conductivity (given in units of $W/cm K$), L_m the linear length of the metallic channel, T_{max} the temperature at the center of the channel, and T_s the temperature of the metallic clusters surrounding the hot spot. Note that far from the hot spot both the metallic cluster and the substrate have the same temperature T_s .

As suggested before, the hot-spot size may be estimated by equating these two contributions, hence

$$\alpha L^2 (T_{av} - T_s) = \frac{\kappa A}{L_m} (T_{max} - T_s). \quad (A3)$$

The two temperatures T_{max} and T_{av} differ only by a numerical factor of order unity, depending on the exact temperature profile. One may thus assume $(T_{av} - T_s) \simeq (T_{max} - T_s) = \Delta T$.

For a fractal, the actual metallic channel is much longer than the linear size of the hot spot, and is given by the anomalous diffusion length scale¹¹

$$L_m = \xi_0 (L / \xi_0)^{2+\theta}, \quad (A4)$$

where ξ_0 is of the order of the grain size and $\theta = 0.8$ in 2D. Substituting L_m in (A3) yields

$$L = (\xi_0^{1+\theta} \kappa A / \alpha)^{1/(4+\theta)}. \quad (A5)$$

Typical values for the above parameters are $\alpha = 2 W/cm^2 K$, $\kappa = 3.5 W/cm K$, $\xi_0 = 20$ nm, and $A = 10 \times 10$ nm². Using these values one finds $L \simeq 100 - 1000$ nm. The anomalous diffusion assumption holds in the regime $\xi \gg L \gg \xi_0$, which is well satisfied in this case; hence the above assumptions are fulfilled.

The estimation of $h(\omega, T)$ is now straightforward: in equilibrium, the injected power P should be equal to the heat flow $J_s + J_m = 2J_s$; hence

$$h = \frac{\Delta T}{P} = \frac{1}{2\alpha L^2}. \quad (A6)$$

Using the above parameters yields $h \simeq 10^9$ K/W.

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