$1/f$ Noise of Granular Metal-Insulator Composites

Joseph V. Mantese
Department of Physics, Cornell University, Ithaca, New York 14853
and
Watt W. Webb
School of Applied and Engineering Physics, Cornell University, Ithaca, New York 14853
(Received 26 August 1985)

The resistivity ρ varies by 7 decades and the intensity of its fluctuations $[S_\rho(f)]_{1\,\mathrm{Hz}}$ varies by 17 decades in composite films of Pt and Al_2O_3 for a wide range of metal volume percent $33\% \leqslant p \leqslant 100\%$ and temperature $7 < T < 300$ K. The power spectral density $S_\rho(f) \propto 1/f^\alpha$ ($1/f$ noise) with $0.8 < \alpha < 1.2$ at all T and p. The normalized noise intensity $[fS_\rho(f)/\rho^2]_{1\,\mathrm{Hz}}$ rises by 10^5 with decreasing p and saturates below a metal-insulator transition, $p \leqslant 50\%$. Results implicate a dual conduction mechanism for composites combining noisy tunneling and quiet metallic paths.

PACS numbers: 72.70.+m, 05.40.+j, 73.60.Dt

The electrical resistivity ρ of granular composites of metal and insulator generally increases by several orders of magnitude as the metal volume fill fraction p is reduced below a critical threshold p_c.[1] For $p \geqslant p_c$, conduction is dominated by charge percolation along continuous metallic filaments, but below p_c it continues by thermally assisted tunneling or hopping conduction between isolated metal islands.[1,2] Substantial resistivity fluctuations characterize metal-insulator composites with the power spectral density $S_\rho(f) \approx 1/f$ ($1/f$ noise), where f is the frequency in hertz.[3]

Current studies of the percolation problem offer models for ρ and the noise intensity $[S_\rho(f)]_{1\,\mathrm{Hz}}$ in composite and discontinuous conductors.[2,4–10] While percolation conduction on a metallic network is analogous to fluid flow in porous media, application of percolation models to electrical conduction in composites requires multiple conduction mechanisms. Measurements of $S_\rho(f)$ as functions of p and temperature T probe conduction mechanisms, test percolation theory calculations, and may define percolation model requirements for real composite conductors.[6,8,10]

Here we report measurements of $\rho(p,T)$ and $S_\rho(f)$ at 1 Hz in platinum-metal–aluminum-oxide (Pt-Al_2O_3) composites for a wide range of p and T. We found $1/f$ noise for all p and T. Tunneling processes are implicated in the fluctuations of the conductivity, with the metallic paths acting to shunt these fluctuating paths. Therefore, we find that percolation models for these composites should include both tunneling and metallic links as alternative conduction paths to account for both the conductivity and $1/f$ noise.

Stable thin-film conductors of thickness $t \approx 1500$ Å were deposited onto hot (400°C) single-crystal sapphire substrates (bearing Au-Cr contact pads) by coevaporation of Pt and Al_2O_3 in a dual-electron-beam–vacuum system. p was determined by monitoring of the deposition on quartz oscillators. To configure the specimen geometries the films were ion milled to the form of standard four-probe resistors plus a center tap with resistor length $l \approx 40\ \mu$m and width $w \approx 2\ \mu$m.

These five-probe samples were connected into a Wheatstone bridge with the specimen forming two adjoining legs (r_1, r_2 with $r_1 \approx r_2$) and the center tap grounded. The opposite legs consisted of large ($R \geqslant 10 r_1$) wire-wound low-inductance resistors. If $r_1 < 1\ \mathrm{k}\Omega$, the bridge was excited with a 500–2000-Hz sine wave and balanced. The fluctuating balance error signal (measured at the voltage contacts) was amplified by a PAR 116 preamplifier and fed to a PAR 124A lock-in detector. The power spectral density of the lock-in output was measured with an HP 5420A spectrum analyzer. If $r_1 > 1\ \mathrm{k}\Omega$, dc excitation of the bridge was used to avoid stray-capacitance problems and the error signal was simultaneously amplified by two PAR 113 preamplifiers in parallel and the cross power spectral density was measured to reduce preamplifier noise. Details are presented elsewhere.[11]

The power spectral density of the resistivity fluctuations, $S_\rho(f)$, is defined in terms of the observed voltage fluctuations, $S_\rho(f) = (wt/l)^2 S_V(f)/I^2$; and for *all* samples it is found to be of the form $S_\rho(f) \propto 1/\Omega f^\alpha$ with $0.8 \leqslant \alpha \leqslant 1.2$. Here Ω is the sample volume and I the current.[12] The approximate inverse-frequency dependence was observed over two to five decades for all samples at all p and T with no systematic dependence upon either p or T.

Figure 1 presents logarithmic plots of the T dependence of $\rho(T)/\rho(300)$ and a tabulation of $\rho(300)$ for seven composites.[13] For $p > 59\%$ ρ falls linearly with temperature until limited by impurity scattering as in dirty metals. The resistivities at low metal fractions ($p \leqslant 50\%$ Pt) increase as the temperature is reduced as is expected of thermally assisted tunneling conduc-

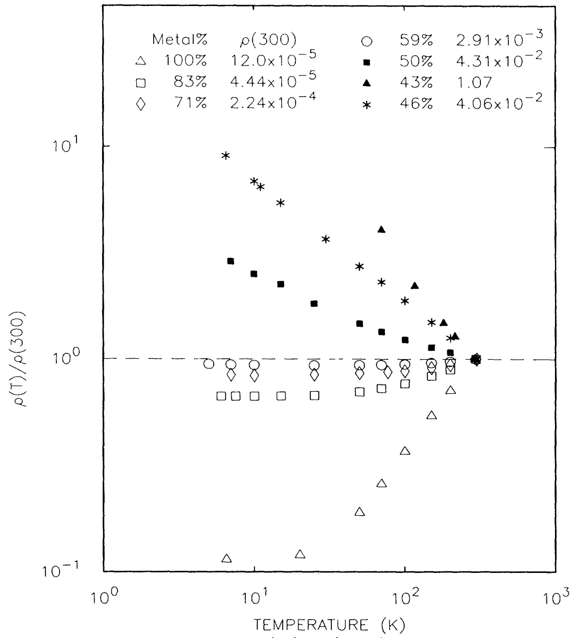

FIG. 1. Logarithm of $\rho(T)/\rho(300)$ as a function of $\log T$ for seven samples, six containing $43\% \leqslant p \leqslant 100\%$ Pt and one with 46% Mo in Al_2O_3. $\rho(300)$ is tabulated in units of ohm centimeters. The dashed horizontal line $d\rho/dT=0$ separates the metallic and hopping regimes.

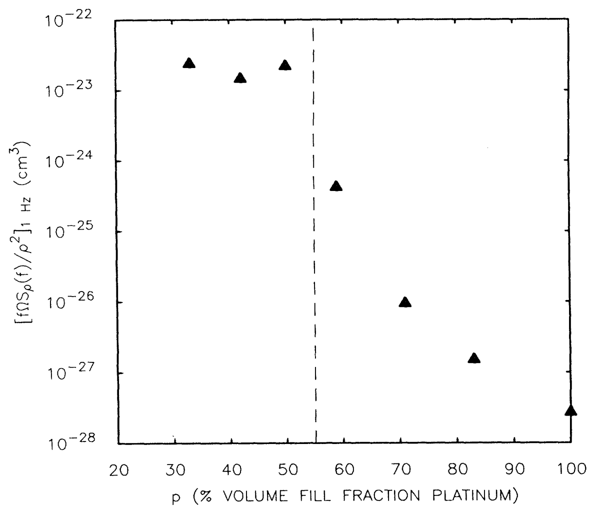

FIG. 2. Normalized noise intensity as a function of p. Saturation occurs for $p < p_c$.

tion in metal-insulator composites.[1,2] If $p < p_c$, no continuous metallic backbone exists, only isolated metal islands or metal strings that decrease in size with p.[1] The transition from metallic conduction to thermally assisted tunneling occurs for p between 59% and 50% Pt.

In Fig. 2 we have plotted the logarithm of the resistivity-normalized spectral intensity, $[f\Omega S_\rho(f)/\rho^2]_{1\ \mathrm{Hz}}$, as a function of p for $T=300$ K. The definition of the resistivity-normalized spectral intensity scales out the inverse-volume and -frequency dependence. From the exceptionally low noise level of pure Pt,[14] $[f\Omega S_\rho(f)/\rho^2]_{1\ \mathrm{Hz}}$ increased monotonically by 5 orders of magnitude with decreasing p and then saturated at p_c where the conduction mechanism changed from metallic conduction to thermally assisted tunneling.

We model the composite conductors as a combination of tunnel junctions and continuous or quasicontinuous metallic paths, the length of the continuous metallic paths varying with p. Qualitatively, composites are represented by a parallel resistor network with two resistances R_T and R_M; R_M represents the conduction through the continuous metal with negligible conductivity fluctuations, and R_T represents the collection of tunneling conduction paths to which we attribute most of the conductance fluctuations.[15,16] From such models we can qualitatively understand the saturation in $S_\rho(f)/\rho^2$ or $S_R(f)/R^2$. The power spectral density of the resistance fluctuation in $R=R_TR_M/(R_T+R_M)$, the parallel combination, is given by $S_R(f)/R^2=\{[R_M/(R_T+R_M)]^2\}S_{R_T}(f)/R_T^2$. For high-metal-content samples $R_M \ll R_T$ and hence $S_R(f)/R^2$ is reduced by the small factor $[R_M/(R_T+R_M)]^2$. As p is reduced, the continuous metallic paths become more tenuous, eventually leaving only finite strings and metal islands when $p < p_c$. Thus as $p \to p_c$, $R_M \to \infty$ and $S_R(f)/R^2$ saturates at the level S_{R_T}/R_T^2. For $p > p_c$ the continuous metallic paths serve to short out some of the conductance fluctuations of the tunnel junctions without themselves adding significantly to the observed noise. For $p < p_c$ both $S_\rho(f)$ and ρ^2 are dominated by the same conduction mechanism, and scale similarly with p leading to saturation of $S_\rho(f)/\rho^2$.

The properties of the tunneling conduction paths are further manifested in the temperature dependence of $S_\rho(f)$ for $p > p_c$. In Fig. 3 $[S_\rho(f,T)/S_\rho(f,300)]_{1\ \mathrm{Hz}}$ is plotted as a function of temperature for $p > p_c$. The normalization scales out the 10^6 variation of $S_\rho(f,300)$. Plotted for comparison are data for an individual Al-Al_2O_3-Al tunnel junction.[15] The striking similarity suggests that the temperature dependence of $S_\rho(f,T)/S_\rho(f,300)$ for $p > p_c$ reflects the conductance fluctuations in the tunneling paths.

Figure 3 also shows that $S_\rho(f,T)/S_\rho(f,300)$ for $p=71\%$ Pt and $p=59\%$ Pt rises rapidly at very low T. The inset shows that for $p=59\%$ Pt, $\rho(T)$ increases approximately logarithmically with T for $T<5$ K as expected for weak localization in two dimensions.[17] Electron-impurity scattering in the metallic strings at

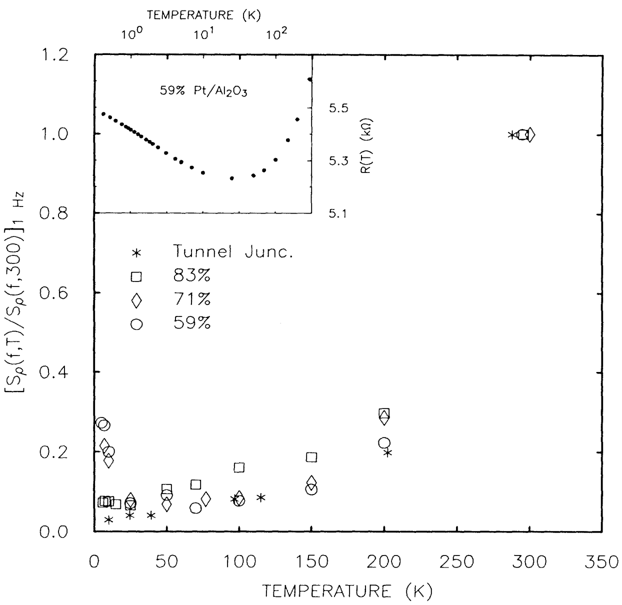

FIG. 3. T dependence of the relative noise intensity for $p > p_c$ and for an individual Al-Al_2O_3-Al tunnel junction. The inset shows an approximate logarithmic dependence of $\rho(T)$ for $p = 59\%$ Pt.

low temperatures should generate such an enhancement in both $\rho(T)$ and $S_\rho(f,T)$.[17,18] Kirkpatrick and Dorfman[18] have predicted that $S_\rho(f,T) \approx 1/Tf$, for a two-dimensional (2D) system; however, it is not clear how their analysis could extend to very low frequencies.

For $p < p_c$, $[S_\rho(f,T)/S_\rho(f,300)]_{1\ \mathrm{Hz}}$ increases rapidly with decreasing temperature as the log-log plots of Fig. 4 illustrate. For $p = 50\%$ the noise magnitude initially decreased by a factor of 2.8 and then rose by more than 2 orders of magnitude; for $p = 43\%$ there is no initial decrease but an even more dramatic rise as T is decreased. Also illustrated are results from another system composed of 46% Mo in Al_2O_3 which shows a similar behavior.

When $p < p_c$ the composite systems are composed primarily of metallic islands imbedded in Al_2O_3 with no continuous metallic backbone. The temperature dependence of the $1/f$ noise then involves several T-dependent factors. Hopping or thermally assisted conductance G_{ij} between islands is expected to involve the product of the tunneling probability from island i to island j, and an Arrhenius factor for the probability of charge transfer requiring an energy $E_{ij} \approx e^2/2Kd$. Here e is the electronic charge, d is on the order of the island diameter, and K is the dielectric constant of the insulator. The island diameter d_i and separations s_{ij} are not uniform but have some, often log normal, distribution. Thus the tunneling conductance represents a sum over terms of the exponential form G_{ij}

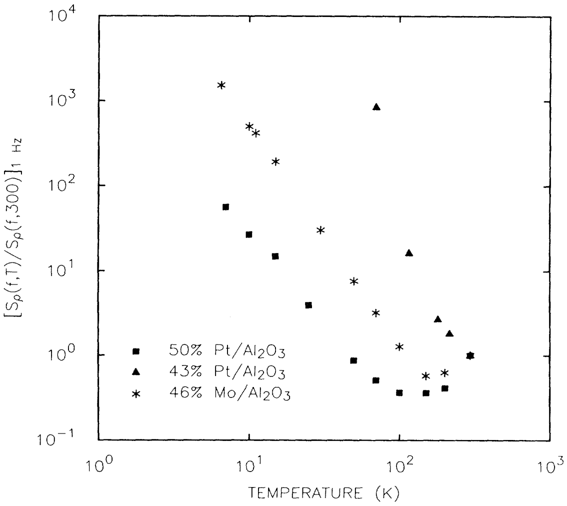

FIG. 4. T dependence of the relative noise intensity for $p \leq p_c$.

$\approx \exp(-2\chi s_{ij} - E_{ij}/k_B T)$, with $E_{ij} \approx E_c$, k_B Boltzmann's constant, and χ representing the tunneling barrier height.[1,2] As the temperature is reduced the probability for charge transfer along some tunneling paths decreases, diminishing the effective number of conducting pathways. Thus $S_\rho(f)$ should rise rapidly with decreasing temperature. However, $S_\rho(f)$ from a single tunnel junction decreases slightly upon cooling. The observed temperature dependence in Fig. 4 reflects the competition between these processes. Note the contrast with Fig. 3 where $p > p_c$. There the Arrhenius factors are suppressed because most metal particles are attached in strings so that E_{ij} is negligible.

Recent theoretical analysis of conduction on random percolation networks of noisy resistors has generated exponents for the resistance and the $1/f$ noise.[8] "Swiss-cheese" models for continuum percolation conduction on the interstices amongst random holes yield additional sets of critical exponents.[6,10] These models all assume a single conduction process that also generates the noise. In contrast, our results demonstrate that metal-oxide composites involve dual conduction mechanisms with metallic conduction controlling the conductivity and thermally assisted tunneling dominating the noise. An analogous disparity is quite likely to apply to some discontinuous metal-film conductors where interparticle tunneling occurs.[7,19]

Present percolation models may nevertheless be directly applicable to metal-insulator composites over a wide composition range under certain conditions and may account qualitatively for $S_\rho(f,T)$ for our composite systems when $p < p_c$ as follows: If the temperature is sufficiently low the temperature dependence of $S_\rho(f,T)$ is dominated by the Arrhenius factors which freeze out conduction paths. Then the variation in magnitude of $S_\rho(f,T)$ due to thermal activation again becomes a percolation problem where variation of p is approximately replaceable by variation in T. Thus the measurements of $\rho(p)$, $S_\rho(p)$, and $\rho(T)$ imply the function $S_\rho(T)$ provided that microstructural similarity is retained. Experimentally $S_\rho(T)$ and $\rho(T)$ are expressible as power laws in T. Thus a scaling law $S_\rho(T) \approx \rho(T)^Q$ can replace the customary $S_\rho(p) \approx \rho(p)^Q$. Lattice theory[8] predicts that $S_\rho(f,T) \approx \rho^Q$, with $Q=2.86$ in 2D and $Q=2.77$ in 3D. For the Swiss-cheese continuum model, Tremblay, Feng, and Breton[20] obtained $Q=5.2$ and $Q=4.1$ in 2D and 3D, respectively, while Garfunkel and Weissman[10] obtained $Q=6.2$ for one version of a 2D Swiss-cheese model. We find that $Q \approx 6.22 \pm 0.06$ for $p=50\%$ Pt and $Q \approx 4.39 \pm 0.04$ for $p=46\%$ Mo by fitting the data in Figs. 1 and 4 over more than a decade in T. The critical-path method[21] implies that effectively $p \approx p_c$ over a wide range of T.

We have found that metal-insulator composite conductors involve multiple conduction mechanisms each distinguished by different noise levels. Therefore, we point out the need for theoretical analysis of dual-conduction-mechanism percolation problems. In our system, the principal features of each mechanism are identifiable with the help of the T and p dependence of ρ and $S_\rho(f)$. The association of p and T dependence allows simple percolation models to be applied approximately for $p < p_c$ and low T. Our results experimentally confirm the common theoretical assumption of local microscopic sources for the $1/f$ noise with the percolation network merely moderating the bulk noise intensity. Finally, we should note that none of these concepts yet identifies the microscopic mechanisms for generation of $1/f$ noise.

We appreciate discussions with Bill Curtin, Dave Wright, Nandini Trivedi, Chris Stanton, André Tremblay, Bob Buhrman, Ping Sheng, and Mark Nelkin, the assistance of John Scofield and Neil Gershenfeld, and the support of the National Science Foundation through the Materials Science Center at Cornell and an individual National Science Foundation Grant, No. DMR-84-14796, to one of us (W.W.W.).

[1] B. Abeles, P. Sheng, M. D. Coutts, and Y. Arie, Adv. Phys. **24**, 407 (1975); B. Abeles, Appl. Solid State Sci. **6**, 1 (1976).

[2] P. Sheng and J. Klafter, Phys. Rev. B **27**, 2583 (1983).

[3] J. V. Mantese, W. I. Goldburg, D. H. Darling, H. G. Craighead, U. J. Gibson, R. A. Buhrman, and W. W. Webb, Solid State Commun. **37**, 353 (1981); J. L. Williams and R. K. Burdett, J. Phys. C **2**, 298, (1969), and Br. J. Appl. Phys. **17**, 977 (1966).

[4] A. L. Efros and B. I. Shklovskii, J. Phys. C **8**, L49 (1975).

[5] J. Klafter and P. Sheng, J. Phys. C **17**, L93 (1984); S. P. McAlister, A. D. Inglis, and D. R. Kroeker, J. Phys. C **17**, L751 (1984); O. Entin-Wohlman, Y. Gefen, and Y. Shapira, J. Phys. C **16**, 1161 (1983).

[6] B. I. Halpern, S. Feng, and P. N. Sen, Phys. Rev. Lett. **54**, 2391 (1985).

[7] M. Celasco, A. Masoero, P. Massetti, and A. Stepanescu, Phys. Rev. B **17**, 2553 (1978).

[8] R. Rammal, C. Tannous, P. Breton, and A.-M.S. Tremblay, Phys. Rev. Lett. **54**, 1718 (1985); R. Rammal, J. Phys. (Paris), Lett. **46**, L129 (1985); R. Rammal, C. Tannous, and A.-M.S. Tremblay, Phys. Rev. A **31**, 2662 (1985).

[9] C. C. Chen and Y. C. Chou, Phys. Rev. Lett. **54**, 2529 (1985).

[10] G. A. Garfunkel and M. B. Weissman, Phys. Rev. Lett. **55**, 296 (1985).

[11] J. V. Mantese, Ph.D. thesis, Cornell University, 1985 (unpublished); J. H. Scofield, Ph.D. thesis, Cornell University, 1984 (unpublished).

[12] We have only confirmed that $S_\rho(f)$ scales inversely with area. Since all samples were approximately 1500 Å thick any normalization by sample volume produces negligible error.

[13] Temperature-dependent data were obtained by placing specimens in a variable-temperature Dewar capable of scanning between 6 and 200 K with 0.5-mK stability below 20 K and 5-mK stability otherwise.

[14] J. H. Scofield, J. V. Mantese, and W. W. Webb, Phys. Rev. B **32**, 736 (1985).

[15] K. Krafft and W. W. Webb, private communication.

[16] C. T. Rogers and R. A. Buhrman, Phys. Rev. Lett. **53**, 1272 (1984), and **55**, 859 (1985).

[17] See the review by G. Bergmann, Phys. Rep. **107**, 1 (1984).

[18] T. R. Kirkpatrick and J. R. Dorfman, Phys. Rev. Lett. **54**, 2631 (1985).

[19] J. E. Morris and T. J. Coutts, Thin Solid Films **47**, 3 (1977).

[20] A.-M.S. Tremblay, S. Feng, and P. Breton, private communication.

[21] V. Ambegaokar, B. I. Halperin, and J. S. Langer, Phys. Rev. B **4**, 2612 (1971).