Nonlinear Behavior near the Percolation Metal-Insulator Transition

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Measurements on discontinuous thin gold films near the percolation threshold show that the current at which nonlinear response takes place, I_{ex}^c, scales with the conductance Σ_0, as $I_{ex}^c \sim \Sigma_0^x$, $x = 1.47 \pm 0.10$. Two possible theoretical models have been considered and studied analytically and numerically. The first is based on local nonlinear contributions from the metallic constituents. The second models nonlinear hopping or tunneling across narrow insulating bridges. Only the second model is compatible with the experiment.

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Continuous phase transitions in inhomogeneous systems are characterized by the vanishing of some physically relevant quantity: the inverse susceptibility (for dilute magnets), the stiffness moduli (for elastically inhomogeneous systems), the conductivity (for metal-insulator mixtures), etc. This is usually accompanied by the shrinkage of the linear-response regime as the transition is approached. In this paper we consider the percolation metal-insulator transition. For metal concentration (p) slightly above the percolation threshold (p_c) and for a sufficiently weak external current (I_{ex}), the voltage (V_{ex}) satisfies $I_{ex} = \Sigma_0 V_{ex}$ where the linear conductance vanishes as $p \to p_c^+$, $\Sigma_0 \sim (p-p_c)^t L^{(d-2)} \sim \xi^{-t/\nu} L^{(d-2)}$. Here ξ is the percolation correlation length and L is the linear dimension of the system. As the external current is increased one may expect deviations from this linear behavior. We define a crossover point (I_{ex}^c, V_{ex}^c) at which the (nonlinear) conductance deviates significantly (by a fraction ε) from Σ_0, i.e., $I_{ex}^c = \Sigma V_{ex}^c$, $\Sigma = \Sigma_0(1+\varepsilon)$. Note that we consider here a reversible process; as the external bias is reduced we return to the linear-response regime.

We report here for the first time results of both experimental and theoretical studies of the crossover to the reversible nonlinear regime.[1] For two-dimensional samples slightly above the percolation threshold our measurements yield

$$I_{ex}^c \sim \Sigma_0^x, \tag{1}$$

with

$$x = 1.47 \pm 0.10. \tag{2}$$

In order to explain the scaling behavior of the crossover point we have considered two phenomenological models. The first model uses a nonlinear random resistor network (NLRRN) which assumes that the conducting backbone consists of microscopic components. The current-voltage characteristics of each such component contains a small nonlinear contribution (physically this may reflect, e.g., local heating effects due to the applied current). The nonlinearity is amplified because of the tenuous structure of the infinite conducting cluster at $p \gtrsim p_c$ and we find for $d = 2$ that

$$x < \nu/t \simeq 1.03. \tag{3}$$

This is incompatible with (2).

The second model is of a dynamic random resistor network (DRRN). This model assumes that in the presence of a sufficiently strong local field a nonconducting channel may become conducting. (This may be used to model the onset of nonlinear hopping or tunneling across narrow insulating "bridges.") We have studied this model in several ways: (i) By employing general scaling arguments we have shown that

$$x < 1 + \nu/t \sim 2.03. \tag{4}$$

(ii) We have performed a finite-cell renormalization-group analysis. This yielded at $d = 2$, $x \simeq 1.68 \pm 10\%$. (iii) By assuming some general analytic properties of $\Sigma(V_{ex})$ we present arguments that suggest $x = 1.5$, in very good agreement with the experiment. Interestingly enough, although we have included a free parameter (α, see below) in our NLRRN, that model could not account for the measured value of x. Thus, by working out phenomenological models we have not only succeeded in explaining the experimental value of x, but have also obtained a strong indication about the microscopic mechanism that governs the nonlinear regime (i.e., nonlinear hopping or tunneling). We hope that our work will stimulate further direct studies of the microscopic aspects of the problem. We also emphasize that the crossover discussed here is quite general and should have qualitative analogs in, e.g., other metal-insulator transitions,[2] the elasticity problem,[3] etc.

The experimental measurements were performed on thin Au films which were near the percolation transition. In previous works the geometry of the systems has been shown to conform with 2D percolation theory[4-6] and some dynamical aspects of these systems, including dc and ac conductivity[7] and $1/f$ noise,[8] have been studied. The fabrication of these films is described in the above references where extensive electron-microscopy studies of the clusters can also be found. However, some important experimental points will be reviewed here.

The Au films, vapor deposited onto a variety of glassy substrates, had a nominal thickness of about 7 nm. Two- and four-terminal electrical contacts were in place on these substrates prior to the evaporation and were used for the room temperature I-V measurements. This experiment required many samples of different resistance values near the percolation threshold. This was accomplished either by closely controlled evaporation techniques in which the samples approach the threshold from the insulator side or by subtractive etching in which they approach from the metal side. Both types of samples exhibited similar nonlinear I-V behavior. At low voltages the samples generally show a linear characteristic as expected and the nonlinearities appear as the bias is increased. It should be observed that at yet higher biases irreversible changes occur. The data reported in this paper were taken in the reversible range.

Next, I_{ex}^c for $\varepsilon = 0.1$ was plotted (see Fig. 1) as a function of $R = 1/\Sigma_0$ and Eq. (2) was obtained from these data. Data were also taken for $\varepsilon = 0.4$ which were indistinguishable from the values at 0.1. The identification of the linear and nonlinear regimes could generally be firmly established by use of computer-generated, point-by-point derivatives of the I_{ex}-V_{ex} curves, i.e., from R-V_{ex} plots.

Our model of NLRRN consists of a bond-percolation system at $p > p_c$. Each bond represents a resistor whose current-voltage relation is given by[9]

$$V = rI - AI^{\alpha}, \tag{5}$$

with $\alpha > 1$. For sufficiently small currents the behavior of each microscopic resistor is basically linear; the crossover to nonlinear behavior is given by $I^c \sim (r/|A|)^{1/(\alpha-1)}$. We are interested, however, in the crossover behavior on the macroscopic scale, L. In general, a combination of microscopic resistors cannot be described by an effective resistor whose behavior is of the form described by Eq. (5). However, if we consider small deviations from linearity, the term AI^{α} can be treated as a small perturbation. Consequently, resistor configurations can be renormalized and we can study the scaling

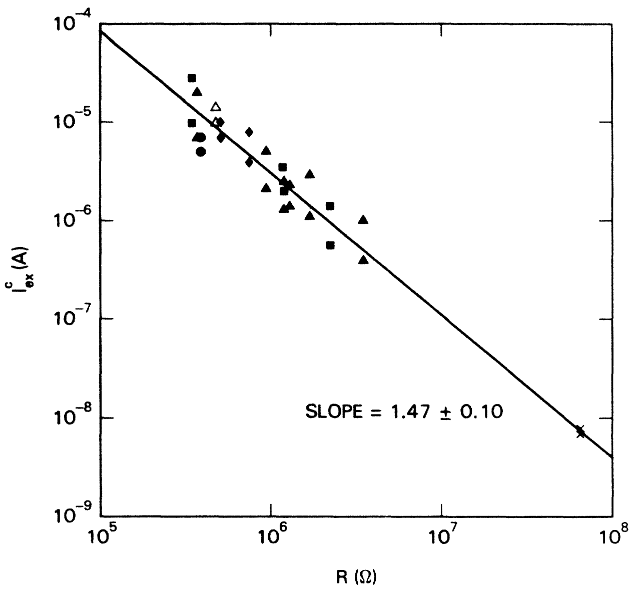

FIG. 1. The crossover current I_{ex}^c vs the resistance R. The points for $\varepsilon = 0.1$ and 0.4 fell on top of each other. The different symbols represent separate fabrication runs with several samples measured from each run. The separation in current at the same resistance is a representation of the uncertainty.

of the parameters r and A. For example, the effective parameters for two identical resistors in series are $r_s' = 2r$, $A_s' = 2A$; for two resistors in parallel we obtain $r_p' = R/2$ and $A_p' = A/2^{\alpha}$. The scaling of I^c is then straightforward.

We have performed calculations on several finitely ramified[10] fractals. We found $I^c(L) \sim \Sigma_0(L)^{-y}$, where $\Sigma_0(L)$ is the linear conductance on the scale L. For topologically one-dimensional structures (resistors in series), $y = 0$. For structures with parallel channels larger macroscopic currents are needed to produce the same effect on the microscopic resistors; I^c scales up and $y > 0$. For the branching triadic Koch curve,[10b] for example, we find that

$$y = [1/(\alpha-1)]\ln[8\times 3^{\alpha}/(2^{\alpha+1}+2+2\times 3^{(\alpha+1)})]/\ln 3.$$

For $1 < \alpha < \infty$, $0.107 > y > 0$. The above considerations imply that $y \geq 0$ for percolation systems,[11] $L < \xi$.

In order to establish the relation to the macroscopic $(L > \xi)$ crossover current, I_{ex}^c, we note that on scales $L \leq \xi$ the number of independent channels is of order 1 and does not scale up with ξ. The current within a block of size ξ is[12]

$$I(\xi) \sim I_{ex}/(L/\xi)^{d-1}. \tag{6}$$

Since $I^c(\xi) \sim \Sigma_0(\xi)^{-y}$ we obtain

$$I_{ex}^c \sim \exp(\{(d-1)v/t + y[(d-2)v/t - 1]\}\ln\Sigma_0)\exp[(d-1-(d-2)\{v/t(d-1)+y[(d-2)v/t-1]\})\ln L],$$

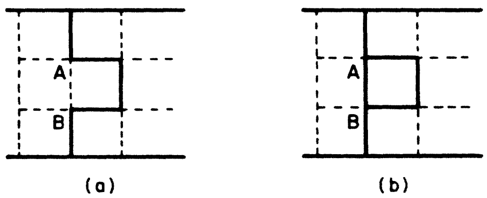

FIG. 2. A 3×3 sample with a conducting path between the electrodes. (a) For an external voltage $V < \frac{5}{3}V_0$ the original bond configuration is maintained. (b) For $V > \frac{5}{3}V_0$ a new conducting channel between A and B is opened.

where $\Sigma_0 \equiv \Sigma_0(L)$. For $d=2$ this yields Eq. (3).

Our second model, DRRN, assumes that once the voltage across an unoccupied bond on the lattice is larger than some critical value, V_0, a conducting bond (identical to an original linear bond) is generated.[13,14] This is demonstrated in Fig. 2.

Within the self-similar scale we shall typically have almost touching tips of conducting clusters. To obtain an estimate for the onset of nonlinearity we first consider a portion of the infinite cluster and assume an extreme case where this cluster forms a one-dimensional loop of size ξ, opened at one end. The voltage drop across that open end is $\Delta V \sim I_{ex}(\xi)/\Sigma_0(\xi) \sim I_{ex}\xi^{1+t/\nu}/L^{d-1}$. The nolinear behavior appears when $\Delta V \sim V_0$, which yields (ignoring factors of L) $I_{ex}^c \sim \xi^{-t/\nu-1} \sim \Sigma_0^{(1+\nu/t)}$. Since in general we may have smaller semiopen loops, as well as parallel channels, we expect inequality (4) to prevail.

Next we have performed a finite-cell renormalization-group calculation. Following Bernasconi[15] we have divided our system into $n \times n$ cells ($n=2$–5). For each cell size we have found the configurational-averaged current-voltage and conductance-voltage curves (see Fig. 3). These curves reflect ~50 h of computer calculation on the VAX 11/780 machine. For $n=2,3$ all possible bond configurations have been averaged over. For $n=4,5$ we have averaged over 10000 configurations each.[16] For each configuration the external voltage was varied starting at V_0 at increments of $\delta(n-1)V_0$, with

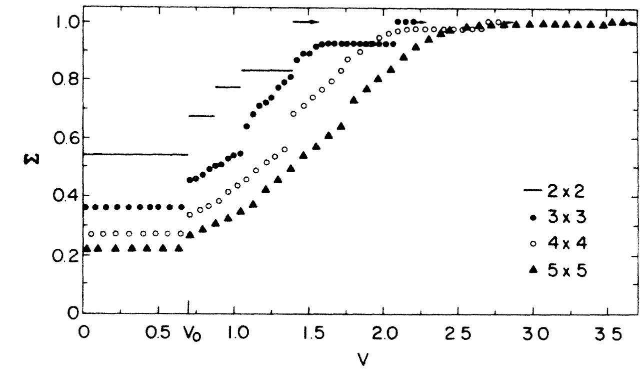

FIG. 3. The conductance Σ vs the external voltage V_{ex} for several sample sizes. The curves are obtained by our taking a geometrical average over random configurations. Arrows indicate the saturated value of Σ (only a fraction of the points is shown).

$\delta = \frac{1}{50}$, and the current distribution was calculated self-consistently. Smaller increments ($\delta = \frac{1}{100}$) did not produce any noticeable change in the curves. Both geometric and arithmetic averaging produced similar results.

For small V_{ex} the conductance approaches its linear value. By finite-size scaling the cell's conductance should scale as $n^{-t/\nu}$ (for $V_{ex} \to 0$). We found $t/\nu \sim 1.0$ in fair agreement (up to ~3%) with the current accepted value. One can also read from Fig. 3 the scaling of I_{ex}^c. The value we find for x is quite sensitive to our choice of ε and the linear rescaling factor. This may suggest that larger cells should be studied before the value of x is determined accurately. For $\varepsilon = 1.6$ we obtained[17] $x \sim 1.68$ with an error in excess of 10%.

The following argument suggests that the value of x may be exactly $\frac{3}{2}$. Setting the conductance of a microscopic resistor equal to 1, Σ reaches a saturation value (n^{d-2}) at $V_{ex} = nV_0$ (at this voltage all channels conduct). One may assume a functional form for $\Sigma(V_{ex})$ which interpolates between its value $\sim n^{d-2-t/\nu}$ at $V_{ex} < V_0$ and the saturation value. If we assume a power-law dependence[18] of the form

$$n^{-t/\nu+d-2} + [(V-V_0)/(nV_0-V_0)]^b (n^{d-2} - n^{-t/\nu+d-2})$$

we find that the crossover occurs at $V_{ex}^c \sim n^{1-t/b\nu}$ ($n < \xi$). For $n > \xi$ we have $I_{ex}^c \sim n^{d-1}\xi^{-t/\nu[(b+1)/b]}$; hence $I_{ex}^c \sim \Sigma_0^{(b+1)/b}$. On the assumption that Σ is an analytic function of V_{ex} for $V_{ex} > V_0$, it should be an even function of the voltage in the limit $V_0 \to 0$. This implies $b=2$ and consequently $x = \frac{3}{2}$ in agreement with the experiment.

Finally we note again that we have shown in the foregoing discussion [e.g., Eq. (6)] that when the external bias (V_{ex} or I_{ex}) is held fixed, the current per channel $I(\xi)$ (on scales $L \sim \xi$) increases as the percolation threshold is approached. This eventually induces nonlinear behavior. Thus, in practically all experimental measurements ($p \to p_c^+$), the nonlinear regime will be reached.

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[1]Note that the problem considered here is different from the nonlinear diffusion problem as discussed, e.g., by M. Barma and D. Dhar, J. Phys. C **16**, 1451 (1983); R. B. Pandey, Phys. Rev. B **30**, 489 (1984); T. Ohtsuki and T. Keyes, Phys. Rev. Lett. **52**, 1177 (1984); Y. Gefen and J. W. Halley, in *Kinetics of Aggregation and Gelation,* edited by F. Family and D. P. Landau (Elsevier, Amsterdam, 1984), p. 161; S. R. White and M. Barma, J. Phys. A **17**, 2995 (1984); Y. Gefen and I. Goldhirsch, J. Phys. A **18**, L1037 (1985).

[2]See, e.g., S. A. Wolf, D. U. Gubser, and Y. Imry, Phys. Rev. Lett. **42**, 324 (1979); Z. Ovadyzhu, private communication. B. Abeles, P. Sheng, M. D. Coutts, and Y. Aire, Adv. Phys. **24**, 407 (1975), have studied nonlinearities in the insulating phase relatively far from p_c.

[3]D. J. Bergman, to be published, has recently worked out scaling relations for the irreversible regime in the elasticity problem; see also P. Ray and B. K. Chakrabarti, unpublished.

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[9]For previous discussions of NLRRN with $r=0$ see, e.g., S. W. Kenkel and J. P. Straley, Phys. Rev. Lett. **49**, 767 (1982); R. Blumenfeld and A. Aharony, J. Phys. A **18**, L443 (1985).

[10a]Y. Gefen, A. Aharony, B. B. Mandelbrot, and S. Kirkpatrick, Phys. Rev. Lett. **47**, 1771 (1981).

[10b]B. B. Mandelbrot, *The Fractal Geometry of Nature* (Freeman, San Francisco, 1982).

[11]For the Berker lattice near p_c with $b=2$ [A. N. Berker and S. Ostlund, J. Phys. C **12**, 4961 (1979)], we find $0<y<0.100$.

[12]Equation (6) has been previously obtained by T. Ohtsuki and T. Keyes [J. Phys. A **17**, L559 (1984)]; see Eq. 22 there. Their analysis (originally applied to superconductor-normal metal mixtures below the superconducting percolation threshold) contains some of the important elements of our NLRRN model. Note, however, that our analysis pertains to $p>p_c$ and contains a nontrivial scaling of $I^c(\xi)$ [see the relation following Eq. (6)]. Our result for the crossover within the NLRRN model (see the expression for I^c_{cx}, which is shown to be incompatible with experiment) is derived here for the first time.

[13]Comparison of V_0 with typical experimental values where the crossover occurs yields $V_0 \simeq 10^3$ V/cm. This value is smaller than the breakdown field and indeed corresponds to reversible nonlinear behavior.

[14]A similar model in the context of dielectric breakdown, $p<p_c$, has been proposed by D. R. Bowman, and D. Stroud, Bull. Am. Phys. Soc. **30**, 563 (1985); P. M. Duxbury, P. Shukla, R. B. Stinchcombe, and J. M. Yeomans, to be published; P. M. Duxbury, P. D. Beale, and P. L. Leath, Phys. Rev. Lett. **57**, 1052 (1986). In the latter it was shown that the voltage at which the first nonlinear event takes place and the voltage at which breakdown of the whole system (irreversible nonlinearity) occurs scale up the same way as a function of concentration. This model (applied to systems with $p<p_c$) differs from our self-consistent DRRN model ($p>p_c$) in mainly the presence of an "avalanche" effect: The first breakdown event makes consecutive events easier. Qualitatively similar avalanche behavior appears also in the random fuse model [see also L. de Arcangelis, S. Redner, and H. J. Herrmann, J. Phys. (Paris), Lett. **46**, 585 (1985)].

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[16]The fluctuations in the calculated value of the conductance did not exceed 1%. This was evaluated by comparison of our results with similar runs over different sets of configurations.

[17]We were led to choose such a high value for ε since for smaller values the response curves, Fig. 3, are not continuous. In the limit of $n \to \infty$ these curves should become smooth.

[18]This behavior may be a special case of a more general behavior where the two asymptotic regimes are governed by two different scaling functions matched at the crossover point. We thank M. E. Fisher for his comments on this point.