Percolation in two-dimensional conductor-insulator networks with controllable anisotropy

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(Received 18 January 1979; revised manuscript received 22 May 1979)

The conductivities of two-dimensional conductor-insulator networks generated photolithographically from laser speckle patterns have been measured. Isotropic networks with $\approx$450 000 statistically independent units show a percolation threshold $f_c \approx 41\%$ conductor and a critical exponent $t \approx 1.30$. Measurements on anisotropic networks and numerical simulations indicate that either f_c, t, or the size of the "asymptotic region" must vary with the degree of anisotropy.

I. INTRODUCTION

Recently, Lobb, Skocpol, and Tinkham[1] and Tinkham[2] have reported electrical-resistivity measurements on a system consisting of superconducting niobium filaments imbedded in a copper matrix, and have interpreted their results in terms of percolation theory. Briefly, they obtained a critical exponent $s = 1.05$, which is different from the value $0.5 \lesssim s \lesssim 0.9$ (Ref. 3) thought to apply to three-dimensional superconductor-resistor systems. They speculate that this discrepancy might be due to the highly anisotropic nature of their system.

Effects in the percolation problem due to this type of anisotropy have also been previously considered theoretically.[4–6] It was the goal of the experiments reported here to examine the effects of systematically introducing anisotropy into a *model* percolating system, in order to obtain a somewhat firmer basis for further theoretical consideration.

To this end, we have invented a system which consists of metal films patterned photolithographically from laser speckle patterns, whose anisotropy is easily varied and controlled. The films are relatively simple to fabricate and contain a large number of statistically independent regions. Moreover, the statistical properties of the speckle patterns which define their geometry have been extensively studied[7] and can be systematically varied.

We find that the effects of introducing anisotropy are nontrivial and fairly dramatic, in that the behavior of the conductance as a function of percent metal appears qualitatively changed. Far from the percolation threshold, these effects can be understood, at least qualitatively, by fairly straightforward arguments. The behavior of the conductance in the more interesting region near the percolation threshold is somewhat inconclusive, although it does provide constraints on the possible alternatives, and should serve as a guide to aid in the development of the theory.

II. EXPERIMENTAL TECHNIQUE

In our experiment, a cw argon-ion laser forms an approximately elliptical and Gaussian spot on a diffuse scatterer (see Fig. 1). A speckle pattern, temporally constant but varying spatially in a pseudorandom fashion, is formed by the scattered light. Its autocorrelation function is related to the shape of the spot; in this case, it is approximately Gaussian with characteristic widths $l_{x,y} \approx \lambda L / a_{x,y}$, where λ is the wavelength of the light (514.5 nm), L is the distance from the spot to the observation plane ($\simeq 1$ m), and $a_{x,y}$ are the widths of the spot ($\simeq 1$ cm).

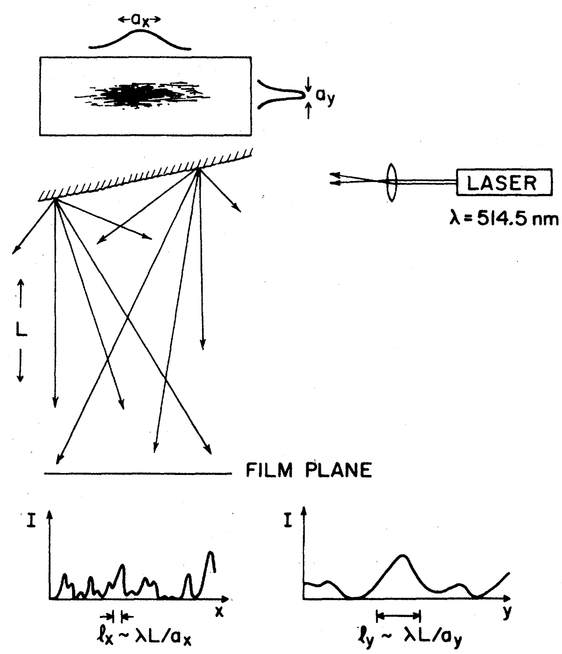

FIG. 1. Schematic of the optical system used to create the speckle patterns. The y direction is out of the plane of the figure.

This pattern exposes extremely high contrast, fine-grained 35-mm film (Kodalith). After development the film contains a pattern of clear and opaque regions whose relative area fractions can be varied by adjusting the laser intensity and the exposure time [see Fig. 2(a)]. This pattern is then photolithographically reproduced as a 100-Å-thick NiCr film on a standard 1×3-in.^{2} glass slide, using standard contact printing, vacuum evaporation, and lift-off techniques [Figs. 2(b)–2(d)]. An enlarged section of an isotropic film is shown in Fig. 3.

The anisotropic speckle patterns are statistically identical to an isotropic pattern modified only by a change of the length scales. Except for relatively minor imperfections, such as edge roughness, introduced by the photolithographic processing, this should apply as well to the metal film. The degree of anisotropy is characterized by the aspect ratio $\alpha \equiv l_x/l_y$, where x is the direction of overall current flow. α is determined experimentally from the ratio a_y/a_x of the axes of the spot on the scatterer. Direct measurements on the films always give a value for α which is closer to unity; we attribute this to the finite resolution of the 35-mm film. The smaller of $l_{x,y}$, which ranged between 30 and 40 μm, is determined as half the mean distance between metal-glass boundaries along the appropriate direction on a sample with $f \simeq 50\%$, and may be interpreted to be the length of a "statistical unit" in that direction. Figure 4 shows an enlarged section from an anisotropic sample.

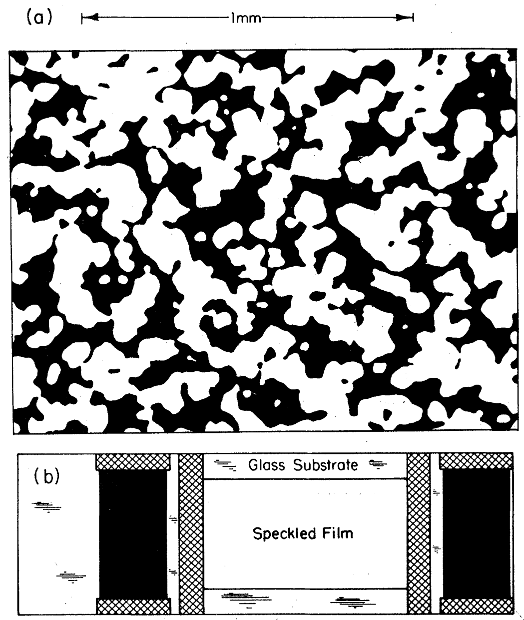

FIG. 3. (a) Photograph of a small section from an isotropic sample: $\alpha=1$, $f=0.419$. The black areas are metal. (b) Sample geometry. The cross-hatched regions are the thick aluminum or NiCr contacts.

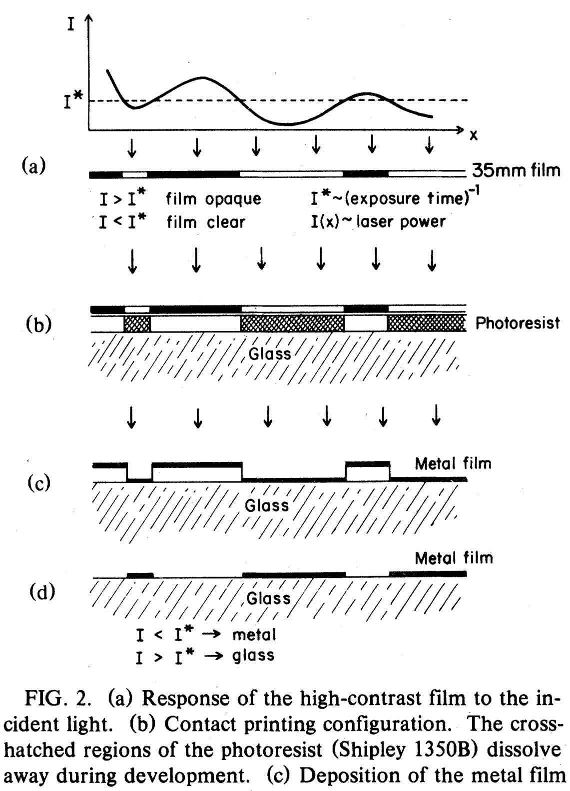

FIG. 2. (a) Response of the high-contrast film to the incident light. (b) Contact printing configuration. The cross-hatched regions of the photoresist (Shipley 1350B) dissolve away during development. (c) Deposition of the metal film by vacuum evaporation. (d) The remaining photoresist is removed by dissolving in acetone, leaving behind a metal film in those regions where the speckle pattern intensity was below the threshold I^*.

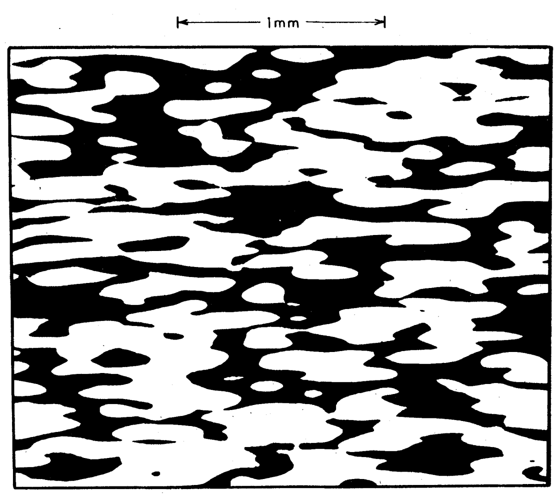

FIG. 4. Photograph of a section from an anisotropic sample: $\alpha=5$, $f=0.407$. The black areas are metal. Note that the magnification is less than that of Fig. 3.

The metallic area fraction f of the film is determined by measuring the optical transmissivity separately for six regions of the film, each comprising $\simeq\frac{1}{8}$ of its area, and interpolating between the measured transmissivities of the reference films ($f=1$) and the bare glass ($f=0$). For the isotropic samples, the measured fluctuations are $\simeq 2\%$, compared with the expected statistical fluctuations of $\approx[nf(1-f)]^{-1/2}/n\approx\frac{1}{4}\%$, where n is the number of statistical units in the sampled area. This nonuniformity appears to be due to either uneven development or intrinsic nonuniformity of the Kodalith film. Great care was needed in the film processing to decrease the nonuniformity to this level.

The uneven processing of the film, its limited resolution, and also other factors such as imperfect optics and nonuniform thickness of the metal film, all combine to make this less than an "ideal" model system. All of these problems can, in principle, be reduced. For example, the films can be made more uniform by increasing the characteristic length scale of the patterns; however, this would be at the expense of increased statistical fluctuations. Photographing the speckle patterns on glass plates, with significantly better resolution, would obtain a similar improvement, but at the expense of costlier and more cumbersome processing. For the purpose of this particular set of experiments, whose primary goal was the qualitative understanding of the effects of introducing anisotropy, we consider these imperfections to be relatively unimportant.

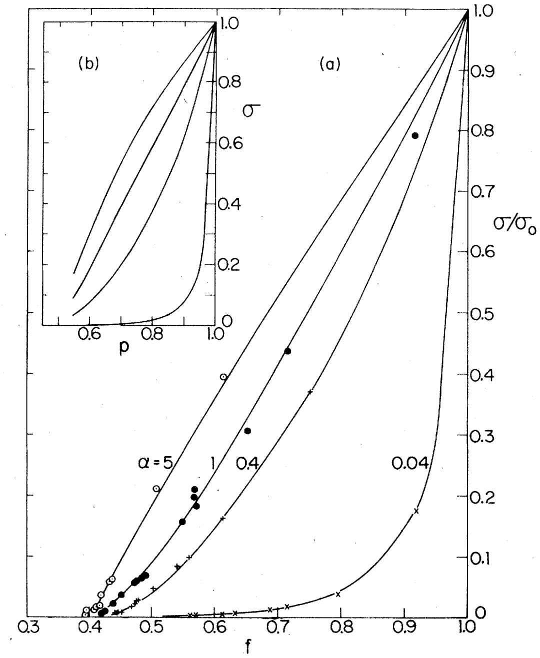

FIG. 5. (a) Normalized conductivity vs metal area fraction. The solid curves are a guide to the eye. (b) Results of Monte Carlo calculations on a square lattice with the same degrees of anisotropy as the data in part (a).

III. RESULTS AND DISCUSSION

Normalized conductivity σ/σ_0 vs area fraction f for samples with aspect ratios $\alpha=5$, 1, 0.4, and 0.04 is plotted in Fig. 5(a) and listed in Table I. The scatterer used was opal glass for the first three sets of samples, and white paper for the fourth. For comparison with a more familiar system, we performed Monte Carlo calculations on a 50 by 50 site conductor lattice using a standard relaxation technique[8]; these results are shown in Fig. 5(b). The conductors in these lattices had equal probability p of being present in both directions, but their values were chosen to be 1 in the direction parallel to the overall current flow, and α^2 in the perpendicular direction. (Our films are electrically equivalent to films with an isotropic geometry, but with local conductivities in the two directions which differ by α^2.) The graphs show very similar variation with anisotropy but they cannot be compared point for point because the microstructures of the networks are different and they have different percolation thresholds. The Monte Carlo arrays contained many fewer conductors than the number of units in the isotropic films, decreasing the statistical accuracy of the numerical results. This difficulty is compounded near p_c, where convergence becomes much slower. For these reasons the Monte Carlo graphs are terminated at $p\approx 55\%$.

The qualitative effects of introducing anisotropy in the conductor lattice can be explained as follows: For any value of $p_c<p<1$, decreasing α^2 will decrease $\approx$half of the conductors in the array, and will thus decrease the net conductance of the array. The conductances for $p\leqslant p_c$ and $p=1$ remain fixed at 0 and 1, respectively. In the limit $\alpha^2\rightarrow 0$, the conductance of the array approaches zero, and the array is effectively one dimensional. In the limit of $\alpha^2\rightarrow\infty$, the conductance will increase towards a finite limiting value, $\sigma(\infty)$. The p dependence of $\sigma(\infty)$ is not known, but a possible upper limit is $B(p)$,[9] the probability of belonging to the backbone of the infinite cluster, since only the backbone conductors contribute to the conductance of the array.

Similar arguments can be applied to the continuum system. Decreasing α at constant f corresponds to making the inclusions short in the direction of overall current flow and proportionately wider in the orthogonal direction. Such an inclusion effectively blocks the current flow over a wider region of the film, so that the conductance of the film as a whole, at fixed area fraction f, will decrease. In this limit, the film is analagous to a sheet of metal foil with long slits cut across it with a razor blade. Because of the large

TABLE I. Metal area fractions f (± 0.002) and normalized conductivities σ/σ_0 ($\pm 1\%$) for each of our samples [see also Figs. 5(a) and 7].

$\alpha=1$		$\alpha=5$		$\alpha=0.4$		$\alpha=0.04$	
f	σ/σ_0	f	σ/σ_0	f	σ/σ_0	f	σ/σ_0
0.916	0.792	0.613	0.395	0.751	0.370	0.918	0.175
0.715	0.436	0.507	0.211	0.613	0.163	0.795	0.0385
0.652	0.306	0.437	0.0624	0.559	0.098	0.714	0.0167
0.571	0.182	0.432	0.0581	0.540	0.0840	0.687	0.0122
0.569	0.211	0.419	0.0360	0.540	0.0828	0.632	0.007 64
0.567	0.210	0.416	0.0194	0.502	0.0477	0.611	0.005 38
0.566	0.197	0.411	0.0165	0.479	0.0287	0.569	0.004 03
0.549	0.157	0.407	0.0131	0.475	0.0265	0.561	0.002 94
0.490	0.0688	0.395	0.0121	0.473	0.0219		
0.484	0.0650	0.394	0.0040	0.467	0.0179		
0.476	0.0602			0.451	0.0083		
0.474	0.0606			0.445	0.0087		
0.473	0.0568			0.443	0.0063		
0.451	0.0369			0.441	0.0069		
0.438	0.0217						
0.425	0.0096						
0.419	0.0065						

depolarization factor of the slits, even a few can cause a sizeable decrease in the conductance of the film. In the opposite limit $\alpha \to \infty$ the slits are directed *with* the current flow. In this limit their depolarization factor approaches zero, so that the conductance of the film approaches f as $f \to 1$. Further from $f=1$ the slits begin to overlap, and can isolate islands of conductor, so that the conductance must not exceed the percolation probability. $B(p)$ does not have as precise a meaning in the continuum case as it does in the discrete bond or site problem, but nonetheless can be operationally defined as that portion of the film which carries an appreciable amount of current.[10] $B(p)$ should be a fairly good approximation to the conductance of the $\alpha=\infty$ system only near $f=1$, and may have altogether different critical behavior as f approaches f_c.

The isotropic films have lengths and widths of roughly 900 and 500, measured in units of the characteristic length of ≈ 35 μm, and contain more statistical units than any of our anisotropic films. Their percolation threshold f_c and critical exponent t were determined by fitting the data by eye to a straight line of the form $\log(\sigma/\sigma_o) = t\log(f-f_c) + \text{const}$. The best values were $f_c \simeq 0.407$, with $t=1.30$, including data within 15% of f_c, although the fit was equally good with the same parameters if all the data were included. These data are plotted in Fig. 6, together with an estimate due to Straley[11] of

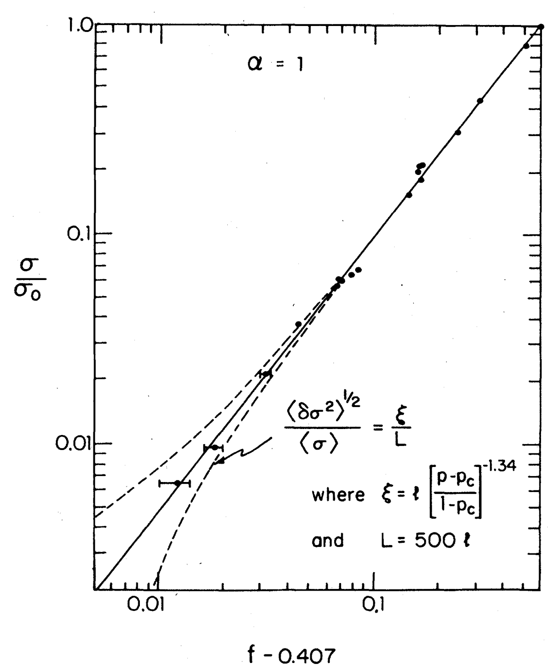

FIG. 6. Normalized conductivity vs $f-0.407$ for the isotropic samples. The dashed curve is an estimate due to Straley (Ref. 11) of the expected fluctuations of the conductance.

the expected statistical fluctuations of the conductivity if the experiment were repeated many times with different speckle patterns of similar statistical properties.

This percolation threshold is an interesting result because it differs from the value $\frac{1}{2}$ which would characterize systems with perfect symmetry between the metal and insulating regions,[12] and because it should be calculable from first principles, at least for an "ideal" speckle pattern for which the intensity distribution is exponential and the other statistical functions are either known or can be derived. Close inspection of a speckle pattern gives the impression that the dark regions — which become metal after the photolithographic processing — look vaguely like channels which separate the bright "speckles". This description is similar to that of a random network of overlapping insulating circles for which $f_c \simeq 0.33$,[13] and suggests that $f_c < \frac{1}{2}$. Of course, this does not apply for all two-dimensional continuous media. By interchanging the metallic and insulating regions, or by using speckle patterns which were made more symmetric by any one of several methods,[7] the percolation threshold ought to be variable over a substantial range on either side of $\frac{1}{2}$. An explanation of the value of the percolation threshold in terms of the fundamental statistical properties of the pattern would be highly desirable.

In the limit of an infinitely large network, we would expect our anisotropic samples to be characterized by the same percolation threshold, because "stretching" the pattern does not change its connectedness. It is well known, however, that fluctuations in a finite sample become large near the percolation threshold, so that different samples may have a conduction onset over some range of values on either side of the "bulk" percolation threshold which characterizes the infinite system.[14] A rough estimate of this range is the condition that the coherence length ξ becomes comparable to the size L of the sample,

$$\xi \sim |p-p_c|^{-\nu} \approx L; \quad \nu \approx 1.35 .$$

If the dimensions of the sample are isotropic, the *expected* value of the conduction onset should not differ appreciably from the bulk value, but if the sample dimensions are anisotropic, the expected conduction onset should be different from the bulk percolation threshold. For our most anisotropic sample with $\alpha = 0.04$, conduction ceases somewhere in the range $51.4\% \leq f < 56\%$. This sample is topologically equivalent to an isotropic sample ≈ 1100 units long and only ≈ 25 units across. Since the probability of finding an insulating path across the width of a finite sample increases as it is made longer and narrower, conduction should cease well above the bulk percolation threshold. Similarly, we expect that some of the $\alpha = 5$ samples, which are ≈ 150 units long and ≈ 400 units across, conduct below the bulk percolation threshold. Again, this is consistent with the observed data.

The statistical sizes of the anisotropic samples are much smaller than the size of the isotropic samples, so that it is more difficult to obtain from them an unambiguous value for the critical exponent. The value obtained depends strongly on the precise choice of the percolation threshold, and on the extent of the "asymptotic region" over which the fit is presumed to be reasonably valid. By making assumptions about one or two of these quantities, information can be obtained about the others. For example, the values $t = 1.30$ and $f_c = 0.407$ for the isotropic data depend somewhat on the assumption that the asymptotic region extends at least 15% above f_c.

If we assume that the anisotropic systems have the same critical exponent and the same asymptotic extent,[15] we find that a good fit can be obtained with the values $f_c(\alpha) = 0.401$, 0.410, 0.427, and 0.500, in order of decreasing α, with $t = 1.24$. This systematic variation is in the same direction as would be expected by considering the conduction onset dependence on the anisotropy of the sample dimensions, although a quantitative comparison is difficult.

A second possibility is to assume that the percolation threshold is the same for all the samples, as would be expected for systems with infinite size. Assuming an asymptotic region which ranges 15% above f_c, we determine a best value of $f_c = 0.410$ and "critical exponents" $t = 0.85$, 1.24, 1.75, and 2.3, in order of decreasing α (Fig. 7). Analysis of the Monte Car-

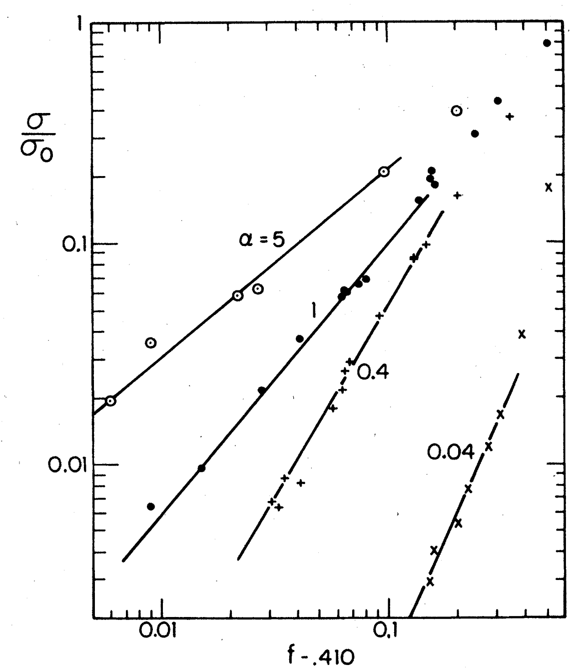

FIG. 7. Normalized conductivity vs $f - 0.410$. The straight lines are fits to the data.

lo data yields similar results. In this case, changing the anisotropy does not alter the sample size. We take $p_c = \frac{1}{2}$, and fit between $0.55 < p < 0.65$ to obtain "exponents" of 0.8, 1.0, 1.2, and 1.2.

These latter results are in disagreement with the assumption of universality, which states that the critical exponents should be independent of the detailed microstructures; in these cases, the aspect ratio of the patterns and the ratio of of the conductances. Moreover, Halperin[16] has pointed out that the conductance of the anisotropic system is bounded above and below by the conductance of the isotropic system, with two values of σ_0 equal to 1 and α^2. It follows rigorously that the asymptotic exponent t for these systems does not vary with α.

We have done renormalization-group calculations in order to try to understand the behavior of the conductivity in the critical region near p_c. We have used a cell introduced previously by Reynolds[17] and Bernasconi,[3] with the modification that the values of the conductors may be anisotropic, as in the Monte Carlo calculations. Our preliminary results indicate that in the limit $p \rightarrow p_c$, for any fixed α (except $\alpha = 0$ or ∞), the macroscopic conductivity of the conductor network becomes isotropic. This reaffirms the earlier conclusions of Bernasconi [4] and Shklovskii.[5] The macroscopic conductivity of our patterned films in the two principal directions must therefore become anisotropic in the ratio $\sigma(\alpha)/\sigma(1/\alpha) \rightarrow \alpha^2$ as $f \rightarrow f_c$, since they can be modeled by an anisotropic conductor lattice after application of a length scale transformation, which also changes the bulk conductivity by the factor of α. We also find, not surprisingly, that the asymptotic critical exponent is simply the isotropic exponent. However, the size of the asymptotic critical region becomes vanishingly small as the system becomes infinitely anisotropic, so that experimental results of highly anisotropic systems might be expected to give an incorrect value for the critical exponent. This provides a possible explanation for the data of Lobb *et al.*[1] These calculations will be presented in full in a future paper.

To summarize, we find that the asymptotic behavior of our conductance measurements on random inhomogeneous films with different anisotropies can be explained in either of two ways. If we allow f_c to be independently chosen for each anisotropy, it is possible to obtain the same effective critical exponent by fitting over a range of f extending $\sim 10-15\%$ above f_c. Alternatively, if we assume that f_c remains fixed as the anisotropy is varied, we obtain an effective critical exponent which depends on the anisotropy. Since the asymptotic critical exponent must be independent of anisotropy, this suggests that it is the extent of the asymptotic region which depends on anisotropy. This latter interpretation is supported by our numerical simulations and by our renormalization-group calculations.

ACKNOWLEDGMENTS

We wish to thank Professor B. I. Halperin for several useful discussions about percolation and the extent of the asymptotic region, and particularly for his observation that the *asymptotic* critical exponent should not depend on anisotropy. We thank him and Professor W. J. Skocpol for their critical readings of this manuscript. This work was supported in part by the NSF and the Harvard Materials Research Laboratory.

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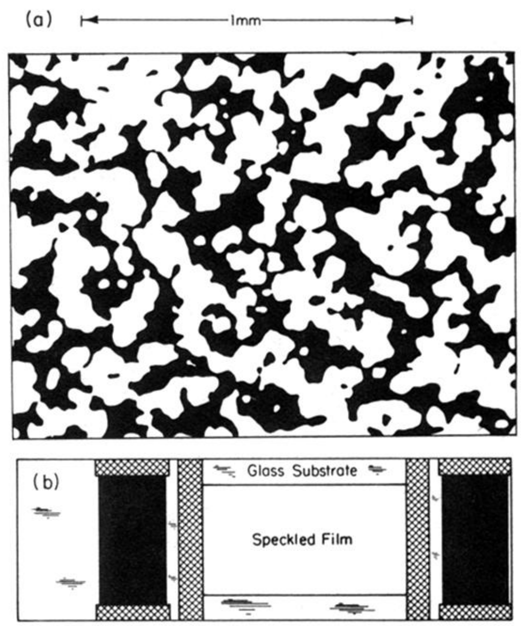

FIG. 3. (a) Photograph of a small section from an isotropic sample: $\alpha=1$, $f=0.419$. The black areas are metal. (b) Sample geometry. The cross-hatched regions are the thick aluminum or NiCr contacts.

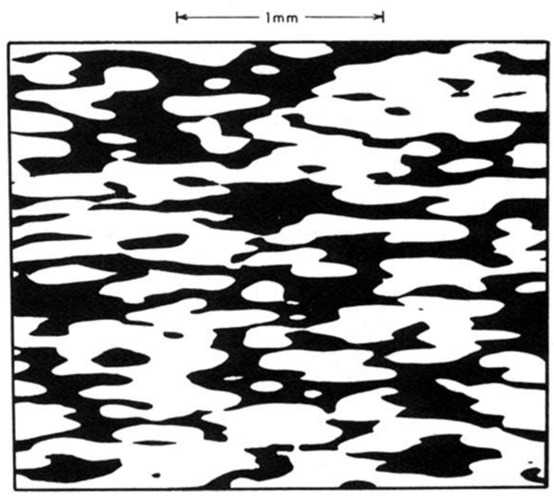

FIG. 4. Photograph of a section from an anisotropic sample: $\alpha = 5$, $f = 0.407$. The black areas are metal. Note that the magnification is less than that of Fig. 3.