Nonuniversal breakdown behavior in superconducting and dielectric composites

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We discuss the critical current density J_c of composites where a fraction p of the material is superconducting and the dielectric breakdown electric field E_c of composites where a fraction p of the material is metallic. Both of these nonlinear processes are characterized by power-law behavior near the percolation threshold p_c, $J_c \propto (p-p_c)^v$ and $E_c \propto (p_c-p)^y$. The exponents v and y are nonuniversal, depending strongly on both the dimensionality and microstructure of the composite.

I. INTRODUCTION

Percolation theory has been very successful in describing the bulk properties of composite materials.[1] Although a limitless number of different composites can be made, a great deal of attention has been focused on systems in which the components are greatly different, and neither component can be viewed as a small perturbation on the other. There are two reasons for this. The first is that percolative systems with only a small amount of a second component embedded in the majority component, or systems where the properties of the two components are not too different, can be treated with accurate perturbative techniques. The second reason is that, if various scaling assumptions are correct, a knowledge of the behavior when the components are greatly different can be used to explain less extreme cases.[2]

One such extreme case is a metal-insulator mixture in which the metal is continuously connected throughout the sample. The conductivity σ of such a mixture will go to zero as more and more metal is removed. If p is the volume fraction of metal present in the mixture and p_c is the critical volume fraction at which the conductivity goes to zero, then

$$\sigma \propto (p-p_c)^t , \tag{1}$$

where t is the critical exponent characterizing the conductivity. Equation (1) is found to hold for a small range of $p \geqq p_c$.

Other properties have their own critical exponents, viz.,

$$k \propto (p-p_c)^e , \tag{2}$$

where k is the fluid permeability of a material in which p of the volume is open to fluid flow,

$$N \propto (p-p_c)^f , \tag{3}$$

where N is an elastic constant for a material where $1-p$ of the volume consists of holes,

$$J_c \propto (p-p_c)^v , \tag{4}$$

where J_c is the critical current density of a material which has a superconducting fraction p, and

$$E_c \propto (p_c-p)^y , \tag{5}$$

where E_c is the breakdown electric field in a dielectric-metal mixture which has a fraction p of metal.

While it has been known for a long time that the critical volume fraction p_c depends on the morphology of the material, it was widely assumed that the exponents were universal, depending only on the dimensionality. Furthermore, it was generally believed—if seldom explicitly stated—that the quantities such as σ, k, and N (but not J_c or E_c), which all appear in linear constitutive relations, would all have the same critical exponents, i.e., $t=e=f$.[3]

In a series of important papers, these beliefs have been shown to be incorrect. Feng and Sen[4] and Kantor and Webman[5] (KW) showed that the exponents t and f were not the same for lattice percolation. Halperin, Feng, and Sen[6] (HFS) extended the KW analysis to continuum systems and showed that any single exponent, such as t, was not necessarily the same for all systems, but could depend on the morphology as well as the dimensionality.

Various nonlinear phenomena, such as the current-induced destruction of superconductivity or dielectric breakdown, have also been studied in percolating systems.[7–13] In this paper, we will extend the analysis of HFS to study such phenomena in systems with a variety of structures.[11] As will be seen below, these properties are very sensitive to morphology in all dimensions. In addition, the results of our calculations rely on fewer approximations than the results for σ, k, and N, and thus may be a more accurate test of the HFS approach.

II. NODES, LINKS, AND BLOBS

Consider a d-dimensional composite material, a fraction p of which is superconducting, with $1-p$ being nonsuperconducting. The superconducting fraction can be divided into two different parts. First, there are isolated clusters of superconductor which do not, in an infinite system, contribute to lossless transport. If the system has enough superconductor so that $p \geqq p_c$, there will also be an

infinite cluster of superconductor, which carries the supercurrent.

The infinite cluster can be further divided into dangling ends, which can be removed from the cluster by cutting a single segment of superconductor, and the backbone. The dangling ends cannot carry a steady-state direct current, so that only the backbone is important for dc transport.

A solvable approximation for the backbone is given by the nodes, links, and blobs model.[14,15] A link can be removed from the backbone by cutting two bonds somewhere in the link, but not any two bonds. This is because a link may have some multiply connected segments, which are called blobs. Links run between nodes, which can only be removed from the backbone by cutting more than two links. A schematic picture of this structure is shown in Fig. (1a).

This model is made quantitative by identifying the average distance between adjacent nodes as the percolation correlation length $\xi(p)$. Near p_c, $\xi(p)$ diverges as

$$\xi(p) \propto l(p-p_c)^{-\nu} , \tag{6}$$

where l is the size of the smallest segment of superconductor, such as the grain size in a granular system, or the bond size in a lattice model. A multiplicative factor of order unity has been omitted from (6), and additional factors of order unity will be omitted from (7)–(10). The critical exponent $\nu=\frac{4}{3}$ in two dimensions,[16] $\nu\cong 0.88$ in three dimensions,[17] and $\nu=\frac{1}{2}$ for $d\geqq 6$.[15]

In order to solve this model, the random backbone is replaced by a uniform hypercubic lattice where all of the nodes are exactly $\xi(p)$ away from their nearest neighbors, and where all of the links, which are different in the real system, are replaced by typical links.[18] This construction is shown in Fig. (1b). With this approximation, consider a hypercube with "volume" L^d, which has faces of "area" L^{d-1}. If a current is injected into one face, and removed from the opposite face, the critical current for the hypercube will be given by

$$I_c = I_{c1}(L/\xi)^{d-1} , \tag{7}$$

where I_{c1} is the critical current of a typical link, and $(L/\xi)^{d-1}$ is the number of parallel links connecting the two faces. Note that phase coherence between the different links is being ignored in (7).

The d-dimensional critical current density j_c is given by I_c/L^{d-1}, so that (7) gives

$$j_c = I_{c1}\xi^{1-d} , \tag{8}$$

which relates the critical current density of the composite to the critical current of a typical link. The only remaining problem, then, is to calculate I_{c1} for various model systems, which will be done below.

For comparison, it is useful to have the corresponding equation for the conductivity of a random system. A derivation similar to the one above gives[15]

$$j = (\xi^{2-d}/R_1)E , \tag{9}$$

where j is the current per unit "area," i.e., the current flowing into a $(d-1)$-dimensional face of a unit hypercube, R_1 is the resistance of a typical link, and E is the electric field. Comparing (9) to Ohm's law $j=\sigma E$, the d-dimensional conductivity σ is given by[15]

$$\sigma = \xi^{2-d}/R_1 . \tag{10}$$

Equations (4), (6), and (8) can easily be used to find the critical current density of a lattice percolation model. In this model, a lattice is constructed from bonds having critical current i_c, and then a fraction $(1-p)$ of the bonds are cut. In this case, a link will also have a critical current $I_{c1}=i_c$, since it is assumed that at least two segments of each link are singly connected. Thus,

$$v = \nu(d-1) \quad \text{(lattice)} , \tag{11}$$

a result which has been derived previously[7] and is numerically well confirmed.[8,9]

Calculating t is more subtle. Singly connected segments of links may not determine the link resistance R_1; the blobs and nodes may also contribute. If, however, only the singly connected segments are assumed to contribute to the resistance—and this is certainly a reasonable lower bound—(10) becomes

$$t = (d-2)\nu + 1 \quad \text{(lattice)} , \tag{12}$$

which seems to be a good approximation for $d\geqq 3$. In writing down (12), it is assumed that the number of singly connected bonds in a link varies as $(p-p_c)^{-1}$ near p_c, which appears to be exact in any number of dimensions.[14] Note, however, that $t\cong 1.30$ in two dimensions,[19] where (12) predicts $t=1$. [For higher dimensions, multiple connections become less likely, and (12) becomes more accu-

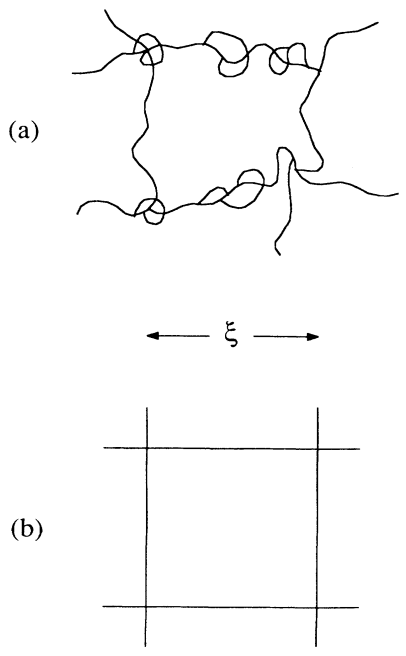

FIG. 1. (a) Schematic picture of a section of the backbone containing four nodes. Upper and lower links contain blobs; left- and right-hand links do not. (b) Solvable model of the backbone, which replaces all of the different links with identical typical links.

rate.[15]] Note, also, that (11) assumes only that there is at least one singly connected bond, while (12) assumes that the resistance comes only from singly connected bonds. Thus, we expect (11) to be more accurate than (12).

III. CONTINUUM SYSTEMS—NODES, LINKS, BLOBS, AND NECKS

Next, consider a uniform d-dimensional superconducting medium which contains d-dimensional spherical nonsuperconducting regions of radius r. These regions, which may consist of either normal metal or insulator, will be called voids. The voids are assumed to occur at random, and can overlap. As more and more voids are created, the system becomes more tenuously connected, and eventually stops being superconducting. The geometrical properties of this system are described by (6), where p is the volume fraction of superconductor, l is a length of order the void radius r, and ν has the same d-dependent values as in the lattice case.[20]

The transport properties of this model "Swiss cheese" system are not the same as the lattice model, however. Instead of being constructed of uniform bonds with critical current i_c, links now have a critical current equal to the critical current of their narrowest segment, where voids have almost cut the link. [See Fig. 2(a).] Assuming a large penetration depth, the critical current of such necks is given by

$$I_{c1}=j_c(\delta_{\min})^{d-1} , \tag{13}$$

where j_c is the d-dimensional critical current "density" of a small cross section of the material (measured in $\mathrm{A/m}^{d-1}$), and $\delta_{\min}$ is the typical minimum neck size of a link. HFS have shown that

$$\delta_{\min}=r(p-p_c) , \tag{14}$$

where a factor of order unity has been ignored. Equations (6), (8), (13), and (14) give

$$J_c=j_c(p-p_c)^{(\nu+1)(d-1)} \tag{15}$$

for the critical current density of Swiss cheese. Comparison to (4) gives

$$v=(\nu+1)(d-1) \quad \text{(Swiss cheese)} . \tag{16}$$

Note that v for this continuum case is different from v for the lattice (11) for any $d\geq 2$.

The conductivity exponent t for the Swiss cheese model has been discussed elsewhere.[6] In two dimensions, the presence of necks does not change the exponent t. For $d\geq 3$ (strictly speaking, $d\geq\frac{5}{2}$), ignoring the contributions of the blobs,

$$t=(d-2)\nu+d-\tfrac{3}{2} \quad \text{(Swiss cheese, } d\geq\tfrac{5}{2}) , \tag{17}$$

which, is once again, different from the lattice expression (12).

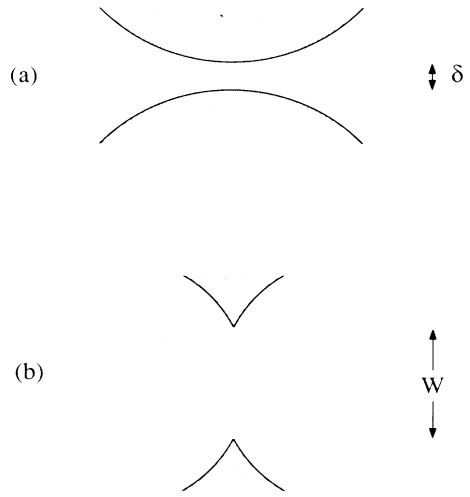

FIG. 2. (a) A neck in the Swiss cheese geometry. Dark areas are holes which come within a distance δ of overlapping in the superconducting case. In the dielectric case, dark areas are metal. (b) The same for inverse Swiss cheese, where the neck width is W.

Next, consider "inverse Swiss cheese." In this structure, a nonsuperconducting matrix contains randomly placed spherical grains of superconductor which can freely interpenetrate. Once again, the critical current of a link will be the critical current of the link's weakest segment, so that

$$I_{c1}=j_c(W_{\min})^{d-1} , \tag{18}$$

where $W_{\min}$ is the smallest neck size in a link, as shown in Fig. 2(b). HFS have shown that, within a multiplicative factor of order unity,

$$W_{\min}=r(p-p_c)^{1/2} , \tag{19}$$

where p is the volume fraction of superconductor. Thus,

$$v=(d-1)(\nu+\tfrac{1}{2}) \quad \text{(inverse Swiss cheese)} , \tag{20}$$

TABLE I. Values of the exponents v and y as a function of the dimensionality d for various model systems. Values of t are included for comparison.

System	v	y	t
Lattice	$(d-1)\nu$	ν	$(d-2)\nu+1$
Swiss cheese	$(d-1)(\nu+1)$	$\nu+\frac{1}{2}$	$(d-2)\nu+d-\frac{3}{2}$ (for $d>\frac{5}{2}$)
Inverse Swiss cheese	$(d-1)(\nu+\frac{1}{2})$	$\nu+1$	$(d-2)\nu+3-d/2$ (for $d>4$)

which falls between the lattice value (11) and the Swiss cheese value (17). Similarly, HFS have shown that t is unchanged for $d=2$ or 3; a simple extension of their method gives

$$t=(d-2)\nu+3-\frac{d}{2} \quad \text{(inverse Swiss cheese, } d\geq 4) \ . \tag{21}$$

These results are summarized in Table I. Note that the different systems considered here have different values of ν, but not of t, for all $d\geq 2$.

IV. DIELECTRIC BREAKDOWN

Next, we consider a problem in which the volume fraction p of good conductor is less than the critical value p_c at which an infinite cluster of good conductor is formed. In this case, the nonlinear problem of interest is dielectric breakdown. We begin by using duality, which relates this problem to the critical current problem in two dimensions.

The idea of duality[2] has proven to be very useful in obtaining critical exponents. For example, the conductivity σ of a mixture of perfect conductor with infinite conductivity and a metal diverges as

$$\sigma \propto (p_c-p)^{-s} \ , \tag{22}$$

where p is the fraction of perfect conductor; this equation should be compared to (1). Using the duality argument, it can be shown that $s=t$ in two dimensions;[2] this result is not true for other dimensions. Similarly, for the lattice case, the critical current density in a superconductor-normal-metal mixture is dual to the dielectric breakdown field in a normal-metal insulator mixture.[9] Thus, for the lattice case in two dimensions, $\nu=y$, where ν and y are defined in (4) and (5).

To go beyond two dimensions, a more general approach is needed. We consider a d-dimensional composite material, a fraction p of which is a metal and $1-p$ of which is an insulator which breaks down at an electric field e_c. (Note that in both of the cases considered in this paper, p represents the material with the higher conductivity.) Below p_c, the metallic component forms finite clusters which are separated by the insulator. The dielectric is assumed to break down and become conducting when the local electric field exceeds e_c. The critical behavior of the macroscopic breakdown field is presumed to be given by (5).

To estimate the exponent y, we use a picture that is similar to the nodes-links-blobs picture, though less frequently invoked.[21] For $p\leq p_c$, the percolating material can be approximated by an array of nodes separated by the correlation length ξ; each node is the center of a metal cluster of linear size of order ξ. Adjacent clusters are separated by a thin membrane of insulator. At some places, the membrane is as thin as possible, i.e., only one insulating bond separates the metal clusters in the lattice case. These bonds are the "singly disconnected bonds" (SDB's). The number of SDB's diverges as $(p_c-p)^{-1}$ as the percolation threshold is approached from below, just as the number of singly connected bonds in a link diverge as the percolation threshold is approached from above.

We first consider the case of percolation on a lattice. A hypercube of size L^d is subjected to an applied voltage V, and hence an average electric field $E=V/L$. Adjacent clusters of metal, which are, on the average, a distance ξ apart, will thus have a potential difference V_1 between them given by

$$V_1/\xi=V/L \ . \tag{23}$$

Dielectric breakdown will occur at places where the *local* electric field exceeds the critical field of the dielectric e_c. These places are located where the separation between adjacent clusters is the smallest. For lattice percolation, this distance is the lattice spacing l. Thus, as the local electric field V_1/l reaches the value e_c, breakdown occurs. This will happen when the external field E reaches a value E_c given by

$$E_c=(l/\xi)e_c \ , \tag{24}$$

which, when combined with (6), gives

$$E_c=e_c(p_c-p)^{\nu} \ . \tag{25}$$

Comparison of this result with (5) gives

$$y=\nu \quad \text{(lattice)} \tag{26}$$

in *all* dimensions. Note that $\nu=y$ in two dimensions, as predicted by duality.

It is instructive to consider the formula for s which results from the same model. Consider a composite consisting of a volume fraction p of perfect conductor ($\sigma=\infty$) and $1-p$ of a normal conductor. The conductivity of this composite is given by (22) near p_c. In the picture just described, the composite consists of hypercubic networks of perfect conductors, separated from each other by thin membranes of normal conductor. If we assume that the conductance of the membranes is dominated by the SDB's, then[6]

$$s=1-(d-2)\nu \ . \tag{27}$$

This equation is analogous to (12), in that it considers only the singly (dis)connected bonds, and is presumably a lower bound for s. In two dimensions it give $s=t$, consistent with duality, but it underestimates s, predicting $s=1$ instead of $s=1.3$. In higher dimensions, the value of s predicted by (27) becomes progressively less accurate, eventually giving the very unphysical value $s=-1$ in $d=6$, where s is known to be 0.[22] This inaccuracy is probably due to the fact that the conductance between two superconducting blobs is progressively less dominated by the SDB's as d increases. [The hyperarea across which transport occurs between such blobs is of order $\xi^{d-1}=(p-p_c)^{-\nu(d-1)}$, while the number of SDB's varies as $(p-p_c)^{-1}$. To make (27) more useful, the additive factor of 1 can be replaced by a variable, ζ_G, which is bounded below by unity and has the value of 2 when $d=6$.[21]] In contrast, E_c is determined by the *single* point of closest approach between the two clusters, and is probably accurately determined by this model.

We can easily extend the analysis which lead to (26) to

continuum systems. We will first consider spherical grains of dielectric randomly placed in a conducting matrix. In keeping with our convention that p is the amount of the better conducting material, we refer to this as Swiss cheese, although the label is more arbitrary here than in the superconducting case considered above. In this case, the voltage between metallic clusters is still given by (24). Breakdown will once again occur at the place where the separation between the clusters is the smallest. This minimum spacing $W_{\min}$ has the form given in (19). Using the same arguments that lead to (24), the critical applied field is found to be

$$E_c = (W_{\min}/\xi) e_c \ . \tag{28}$$

Using (6) and (19), we obtain

$$E_c = e_c (p_c - p)^{\nu + 1/2} \ , \tag{29}$$

which leads to

$$y = \nu + \tfrac{1}{2} \quad \text{(Swiss cheese)} \tag{30}$$

in any dimension. This result applies to a system consisting of overlapping spherical grains of dielectric embedded in a metal. Note that, for this result to be true, it is necessary to assume that the metal points in Fig. 2(b) are not infinitely sharp. If they were, the breakdown field would be zero for all p.

In the inverse Swiss cheese model, metallic grains that can freely interpenetrate are randomly placed in a dielectric matrix. Breakdown will again occur at places of smallest separation, which have a size $\delta_{\min}$, as given by (14). Substituting $\delta_{\min}$ for $W_{\min}$ in (28) leads to the critical exponent for this model,

$$y = \nu + 1 \quad \text{(inverse Swiss cheese)} \tag{31}$$

in any dimension. Results for the exponent y are summarized in Table I.

V. DISCUSSION

Several features of the results we have obtained are worthy of further comment. One point concerns duality as it is applied to continuum models. In the lattice case, in two dimensions, the critical current and dielectric breakdown exponents are equal to each other because of duality.[9] For the continuum case in two dimensions, the exponent v in the Swiss cheese model is found to equal the exponent y in the inverse Swiss cheese model, and the exponent v of the inverse Swiss cheese model is the same as the exponent y in the Swiss cheese model (see Table I). These relations confirm, within the nodes-links-blobs model, that these problems are dual to each other. Since the duality transformation consists of interchanging superconductor and insulator and interchanging fields and currents, this equivalence of v and y for dual geometries (i.e., Swiss cheese and inverse Swiss cheese) is exact. (For more detailed discussions of duality, we refer the reader to Refs. 23 and 2.) Unfortunately, no such duality relations exist outside of two dimensions.

The results for critical current presented here do not, of course, take into account the full complexity of real normal-superconducting composites. Phase coherence, which will presumably introduce correlations between nearby bonds, has been completely neglected in our analysis. The assumed current-voltage characteristic for a single link has also been simplified, because we have assumed that a link remains superconducting up to some critical current, after which it becomes normal, ignoring the rounding that occurs in real weak links. With these simplifications, our model is similar to a random fuse network, such as recently considered in two dimensions by Arcangelis, Redner, and Herrmann[10]: The critical current at which their fuses burn out is the analog of the superconducting critical current in our model.

Gefen *et al.*[12] have recently studied a model similar to ours for the lattice case, use finite-size scaling at the percolation threshold. Since they are concerned principally with the I-V characteristics at $p = p_c$, it is not clear that their results can be immediately compared to ours.

Duxbury *et al.*[11] have studied the size effects of breakdown in a random fuse network and in a metal-dielectric network. Using the same argument as that proposed by Lifshitz to study band tails in semiconductor alloys, they found that the critical applied electric field for dielectric breakdown decreases as the size of the system increases. In the limit of an infinite system, the critical applied field goes to zero. This result occurs because of the bunching of field lines around metallic inclusions, and is a result of percolation cluster statistics and Laplace's equation. The results of this paper complement Duxbury's analysis in two ways. First, we believe that our exponents should give the concentration dependence near p_c for large samples where the sample size is much greater than the correlation length $\xi(p)$. Our result is thus in agreement with Duxbury *et al.* for fixed size in the case they discuss (lattice percolation), and, in particular, gives the same power law as they obtain for the behavior of their function $a(p)$ in the lattice case.

Whether or not a system fails in the catastrophic way predicted by Duxbury depends on the details of the current-voltage characteristics of single bonds. In a sample containing mathematically sharp superconducting bonds which are hysteretic, and thus irreversible (that is, the current-voltage characteristic shows a discontinuous jump at the critical current) Duxbury's analysis is appropriate. If, however, the current-induced transition into the normal state is continuous and reversible, a nonzero fraction of the bonds will carry their critical current before a voltage appears across the sample. This type of nonlinear current-voltage characteristic is not described by Laplace's equation. In particular, the bunching of current around a normal-metal inclusion is relieved when the local critical current density is exceeded because the effective resistivity of that region will rise continuously to its maximum value as the current is increased. For such systems, we believe that the results presented in this paper are correct without the additional terms derived by Duxbury.

Finally, it should be noted that critical exponents have been measured on real superconducting samples in both two and three dimensions.[7] The measured exponents were closer to the lattice values than the continuum

values. This may be due to the fact that some real samples may have minimum neck sizes determined by their structure, so that neck sizes will not go to zero, as predicted by (14) or (19). When this happens, the lattice values for the exponents are correct close to p_c. Further experimental work is called for here. It is also possible to simulate the essential aspects of continuum systems on a lattice by choosing a distribution of bond strengths to mimic the distribution of neck sizes which occur in a continuum system. Preliminary results of such simulations in two dimensions,[24] using the method outlined in Ref. 9, are in excellent agreement with the results predicted here.

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