PHYSICAL REVIEW B

CONDENSED MATTER

THIRD SERIES, VOLUME 37, NUMBER 6 15 FEBRUARY 1988-II

Theory of dielectric breakdown in metal-loaded dielectrics

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(Received 19 January 1987; revised manuscript received 26 June 1987)

We analyze a simple model of dielectric breakdown in metal-loaded dielectrics. We show that the breakdown electric field in a large, but finite, sample tends to zero in a universal manner as the fraction of metal in the material approaches the percolation threshold. In addition, the average breakdown electric field decreases logarithmically with the linear dimension of the system when the volume fraction of metal, p, is below the percolation threshold, p_c. The average initial breakdown field, E_1, behaves as $E_1 \approx 1/[A(p)+B(p)\ln(L)]$, where L is the linear dimension of the system. The coefficient of the logarithmic term diverges as p approaches the percolation threshold, $B(p)\sim(p_c-p)^{-\nu}$. The exponent ν is the power-law exponent for the divergence of the percolation correlation length as $p\rightarrow p_c$. The breakdown field distribution function $F_L(E)$ has the form $F_L(E)\approx 1-\exp[-cL^d\exp(-k/E)]$, where d is the dimensionality of the system. These predictions are verified by computer simulations of random mixtures of conducting and nonconducting elements. The breakdown process is modeled by allowing the insulating elements to only be able to withstand a fixed voltage difference before they fail and become conducting elements. This process continues until a macroscopic dielectric failure occurs by the formation of a conducting path across the system. We find that the electric field necessary to cause this complete breakdown is the same as the field necessary to cause a single microscopic failure.

I. INTRODUCTION

This paper addresses the problem of dielectric breakdown in metal-loaded dielectrics.[1-5] These materials consist of an inhomogeneous mixture of conducting and insulating components. A good example of a metal-loaded dielectric is the material used by Grannan, Garland, and Tanner[6] in their study of the dielectric constant of a random mixture of 200-Å Ag particles contained in a matrix of KCl. Another example is solid-fuel rocket propellant.[7] A typical solid-fuel rocket propellant consists of a random mixture of aluminum particles (the fuel), ammonium perchlorate particles (the oxidizer), and a rubbery binder which holds the mixture together. Experiments on dielectric breakdown in solid rocket propellants[7] indicate that the presence of the aluminum particles in the material dramatically reduces the external electric field necessary to cause dielectric breakdown of the material. The breakdown field is a sensitive function of the volume fraction of aluminum in the material. The dielectric breakdown of the material proceeds via a series of microscopic failures which lead to a macroscopic breakdown and electrostatic discharge.

In this paper we analyze a simple model of dielectric breakdown in a mixture of metal and dielectric by means of theoretical arguments and numerical solution of Laplace's equations in a random system. Several important results are derived from this analysis. The sensitivity of the dielectric breakdown field on the metal fraction in the material is caused by the formation of metallic percolation clusters in the material. The breakdown field is of order the inverse of the linear dimension of the largest of these percolation clusters. Since the size of the clusters diverges as the volume fraction of metal tends to the percolation threshold value, the breakdown field tends to zero in this limit.

The breakdown field in our model has an intrinsic size dependence. Large samples of material are more sensitive to dielectric breakdown than small samples. This is because the breakdown begins near the critical defect in the system. The critical defect is (roughly) the largest pair of strongly interacting metallic clusters which are oriented parallel to the bulk electric field. The breakdown electric field is of order the inverse of the linear dimension of this defect. Clearly, since the largest defect in a large sample will be larger than the largest defect in

a small sample, the applied breakdown electric field is smaller in larger samples. Our arguments will show that the breakdown field E_1 is of order $1/\ln(L)$ where L is the linear dimension of the sample.[4,5] The distribution function of breakdown electric field strengths $F_L(E)$ has the functional form[5,8]

$$F_L(E)=1-\exp\left[-cL^d\exp\left[\frac{-k}{E}\right]\right]. \quad (1)$$

In Sec. III we derive this form from a simple scaling argument based on the statistics of percolation clusters.[9] This form is similar to the Weibull distribution[10] ordinarily used to fit the distribution function of breakdown and failure problems. The Weibull breakdown distribution function is of the form

$$F_L(E)=1-\exp(-cL^dE^m). \quad (2)$$

A plot of Eq. (1) looks qualitatively similar to the Weibull form (2) if the Weibull exponent, m, is large. However we will show that (1) fits our numerical data much better than the Weibull form (2). This is because the size distribution function for percolation clusters is an exponentially decreasing function of cluster size. If the cluster sizes had a power-law distribution function then the Weibull form would be the appropriate one.

We test all of these theoretical predictions by solving a lattice Laplace's equation for a random network of conductors and capacitors. In two dimensions we use lattice sizes up to $L=100$ and in three dimensions we use lattice sizes up to $L=35$. The numerical results agree extremely well with the theoretical predictions. The breakdown field is a rapidly decreasing function of the metal fraction near the percolation threshold; the average breakdown field is a logarithmically decreasing function of system size, and the breakdown field distribution function (1) fits the numerical results very accurately. It might be noted that, for two dimensions only, this dielectric breakdown problem is dual[2] to the fuse failure problem discussed in Refs. 4, 5, and 8. The dual variable to the breakdown field below the percolation threshold is the critical current density above the percolation threshold.

Similar work on numerical modeling of dielectric breakdown has been done by other authors. In Ref. 3 the authors model dielectric breakdown in an initially homogeneous system. In that work the randomness is put into the choice of which bond on the lattice to fail next. The failure probability is proportional to the local electric field to the power η. This procedure generates a fractal growth of the breakdown path. Our method of only failing the capacitor with the largest voltage drop corresponds to the limit $\eta\to\infty$. Reference 2 describes independent work similar to ours on dielectric breakdown in metal-loaded dielectrics. References 4 and 5 address the problem of critical current failure in a random network of fuses and insulators.

II. LATTICE MODEL

The simple model[1,2,4,5] we use to describe dielectric breakdown in these metal-loaded dielectric materials is shown in Fig. 1. The nearest-neighbor bonds of a lattice are occupied at random by conductors with probability p. The remaining bonds (probability, $1-p$) are occupied by capacitors. These capacitors can withstand a voltage drop up to one volt. If the voltage drop across any capacitor exceeds 1 V, then the capacitor fails irreversibly and becomes a conductor. The probability p is chosen so that p is less than p_c, the percolation thresh-

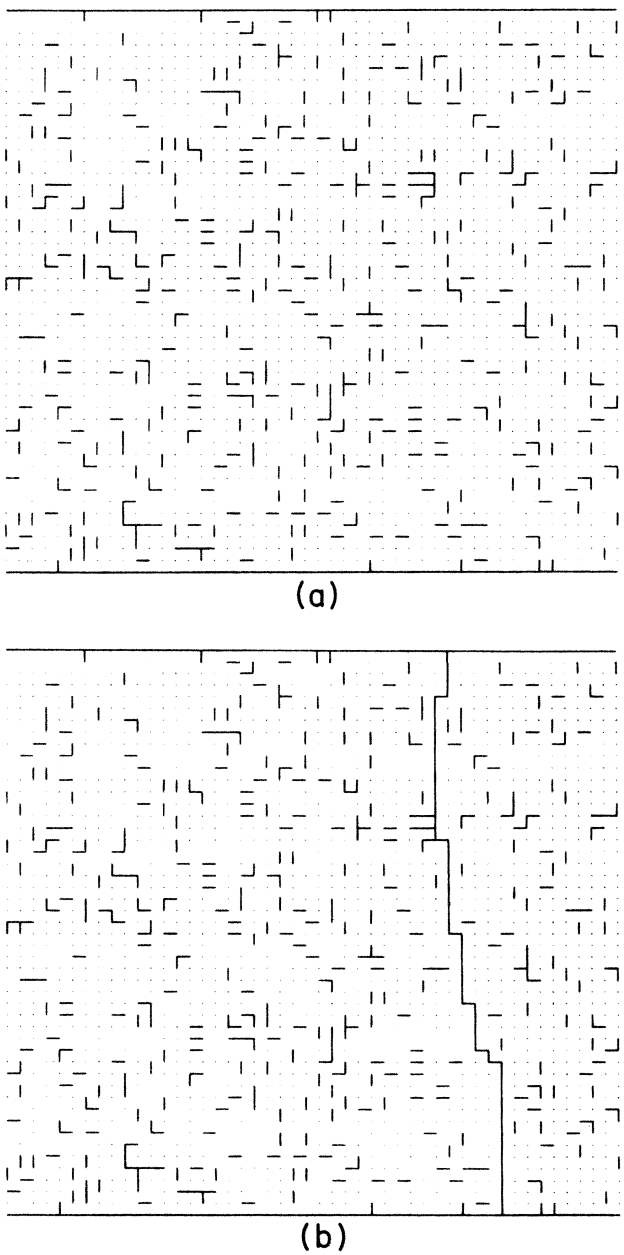

FIG. 1. (a) A typical starting configuration for dielectric breakdown on a two-dimensional square lattice. The top surface is held at voltage V and the bottom surface is grounded. The dots are lattice points and the lines denote conducting paths. Here the metal fraction is $p=0.100$ and the system size is $L=48$. (b) The final configuration after complete breakdown has occurred. Note the conducting path running from top to bottom.

old. This ensures that no conducting path spans the system. This is a simple model of an inhomogeneous material which is composed of a random mixture of metal and dielectric. An external electric field is applied to the model by grounding all sites on the bottom layer of the lattice and fixing the voltage at all sites on the top layer of the lattice to have voltage V.

This model is relevant to an experimental situation in which a sample of dielectric contains a randomly distributed volume fraction of metallic particles. Clusters of metallic particles occur naturally due to their random location in the sample. A large electric field is placed across the sample fast enough so that there is insufficient time for space-charge motion through the (imperfect) dielectric to reduce the electric field in the dielectric. Breakdown occurs in regions of the dielectric where the local electric field exceeds the intrinsic breakdown field of the dielectric.

The initial breakdown of the material is modeled by finding the minimum external voltage V necessary to cause *one* capacitor in the system to fail. We call this initial breakdown voltage V_1. If the lattice has a linear dimension of L then the initial applied breakdown field is given by $E_1 = V_1/L$.

In the pure limit, where all bonds of the lattice are occupied by capacitors, the initial breakdown field is 1 V per lattice spacing. By contrast, in the limit where p is close to the percolation threshold, p_c, the initial breakdown field is very small. This is because for p close to p_c very large conducting clusters exist in the system. For p just below p_c the largest clusters in the system will almost span the system. There will be some capacitors in the system which lie between a cluster attached to the top electrode and a cluster connected to the bottom electrode. In such a circumstance the breakdown voltage will be $V_1 = 1$ V. The breakdown electric field is then $E_1 = 1/L$ volts per lattice spacing. The breakdown electric field will scale like $1/L$ when L is much smaller than ξ, the percolation correlation length. This will occur when p is close to p_c in a large but finite system.

The approximate dependence of E_1 on p can be inferred by a simple argument based on the idea of the critical defect as displayed in Fig. 2. The critical defect is a pair of clusters of metal of total length l oriented along the applied field and separated by a small number of lattice spacings. The electric field between these two clusters is enhanced by a factor of order l times the applied bulk electric field.[11] The probability of finding a percolation cluster of size l is of order $(1/\xi)\exp(-l/\xi)$ where ξ is the percolation correlation length.[9] The largest cluster in a sample of volume L^d is of order $l_{\max} \approx \xi \ln(L^d)$. Since the breakdown field is of order $1/l_{\max}$ the breakdown electric field is

$$E_1 \approx \frac{(p_c - p)^\nu}{\ln(L)} . \tag{3}$$

The dominate dependence is the $(p_c - p)^\nu$ behavior so the breakdown field tends to zero as p approaches p_c. The logarithmic size dependence is less obvious but can be important in very large systems. Note that for any $p < p_c$ the breakdown field tends to zero logarithmically with L and $L \to \infty$.

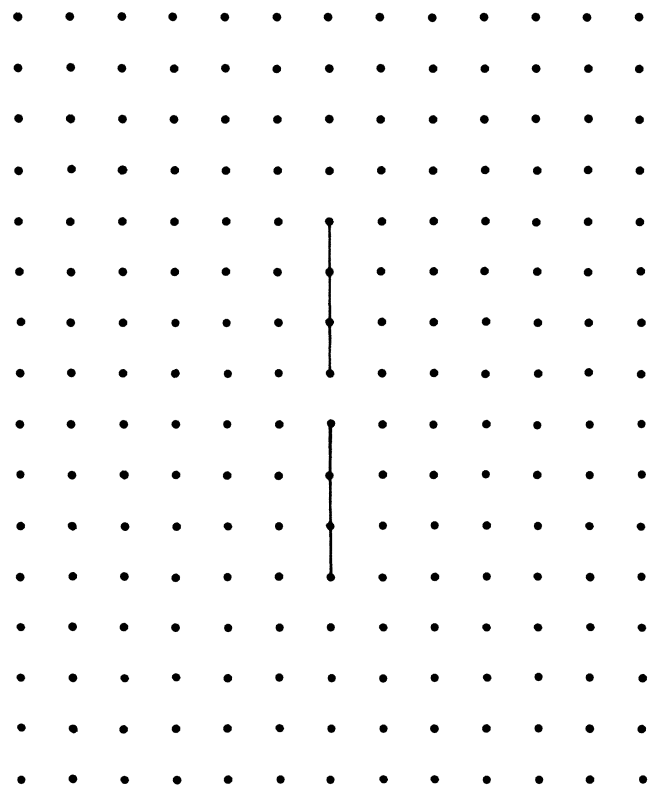

FIG. 2. The critical defect is a pair of roughly linear conducting elements oriented parallel to the applied field. The pictured defect is composed of l connected conducting elements. The breakdown field for this configuration is of order $1/l$. The breakdown will begin between the two conducting clusters.

When the metal fraction is close to zero the metallic clusters will be small. This does not imply that the breakdown electric field tends to unity as $p \to 0$. On the contrary, even if there is only a single defect (conducting element) in the system (as long as it is aligned parallel to the applied field) the breakdown field will be reduced by a finite amount. The problem of a single conducting defect can be solved exactly in terms of the lattice Green's function. In two dimensions the addition of a single conducting defect far from any of the surfaces of the sample will reduce the breakdown field from $E_1 = 1$ V per lattice spacing to $E_1 = \pi/4$ V per lattice spacing. Hence, the addition of even a very small amount of metal to a dielectric material will significantly reduce the initial dielectric breakdown field.

The numerical method we use to solve the lattice Laplace's equation for the random configuration is as follows. The lattice Laplace's equation amounts to solving

$$\sum_{(i,j)} C_{ij}(V_i - V_j) = 0 , \tag{4}$$

where i is summed over all internal sites on the lattice, j

is summed over the nearest neighbors of i, the voltages along the top row of the lattice are set to unity and the voltages along the bottom row are set to zero. The constants C_{ij} are chosen to be $C_{ij}=1$ if sites i and j are connected by a capacitor and are set to $C_{ij}=10^6$ if they are connected by a conductor. This serves to short out sites which are connected by a conductor. The numerical technique used to solve (4) is the conjugate gradient method.[12,13] This method amounts to finding the minimum of the function

$$\frac{1}{2}\sum_{(i,j)} C_{ij}(V_i-V_j)^2 \tag{5}$$

in the $(L-1)L^{d-1}$ dimensional space of the voltages at the internal nodes of the lattice. This method converges reasonably fast and is extremely suitable for vectorization so that the calculation can be done efficiently on a Cyber 205. We relax the system of equations until the residual is smaller than 10^{-15}. On a two-dimensional lattice of size $L=100$ at $p=0.200$ this requires about 14 000 iterations of the conjugate gradient algorithm and takes about 26 s on a Cyber 205. Alternative methods exist for finding the solution to the random lattice Laplace's equation which are more rapidly convergent for p very close to p_c.[13]

III. SIZE DEPENDENCE OF THE BREAKDOWN FIELD

Figure 3 displays a plot of the initial breakdown field E_1 as a function of p for $L=50, 70, 100$ on a two-dimensional lattice. The percolation threshold for this lattice is $p_c=0.50$. The breakdown field E_1 tends to zero as p approaches p_c.

Note that the breakdown field in Fig. 3 is a slowly decreasing function of the system size L. As predicted by Eq. (3), the breakdown field is a logarithmically decreasing function of the linear dimension of the system for large systems. This behavior is demonstrated in Fig. 4. In order to test the logarithmic length scaling argument, we plot the inverse of the average breakdown field versus the logarithm of the system size for several different p's. The data is well described by a linear dependence on the logarithm of the system size. Again, as predicted by Eq. (3), the slopes of the lines are small for p close to zero and large for p close to p_c. From these data we can make the empirical observation that the average initial breakdown field E_1 scales like

$$E_1 \approx \frac{1}{A(p)+B(p)\ln(L)} . \tag{6}$$

By comparing with Eq. (3) the coefficient of the logarithmic term is $B(p)\approx(p_c-p)^{-\nu}$. To test this behavior, Fig. 5 shows a plot of $\ln(B)$ versus $\ln[(p_c-p)/p_c]$. As expected, the power-law fit to the data give $\nu=1.46\pm0.22$ which is consistent with the two-dimensional value[9] $\nu=\frac{4}{3}$.

Since the systems being modeled are random, different samples will have different breakdown fields. However, there does exist a smooth distribution function for the breakdown fields of samples with a given p and L. We

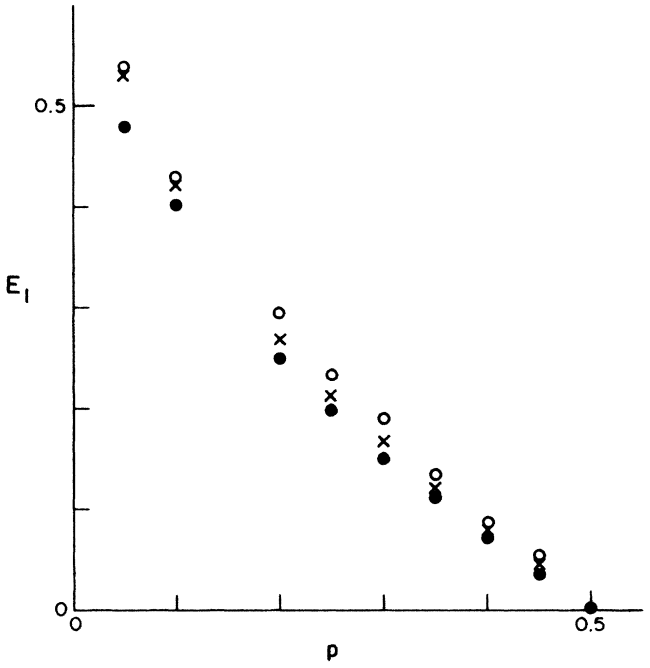

FIG. 3. The initial breakdown field, E_1, as a function of the conducting fraction, p, and the system size L. The breakdown field tends to zero as p approaches the percolation threshold $p=p_c$. Note that E_1 is a slowly decreasing function of system size at fixed p.

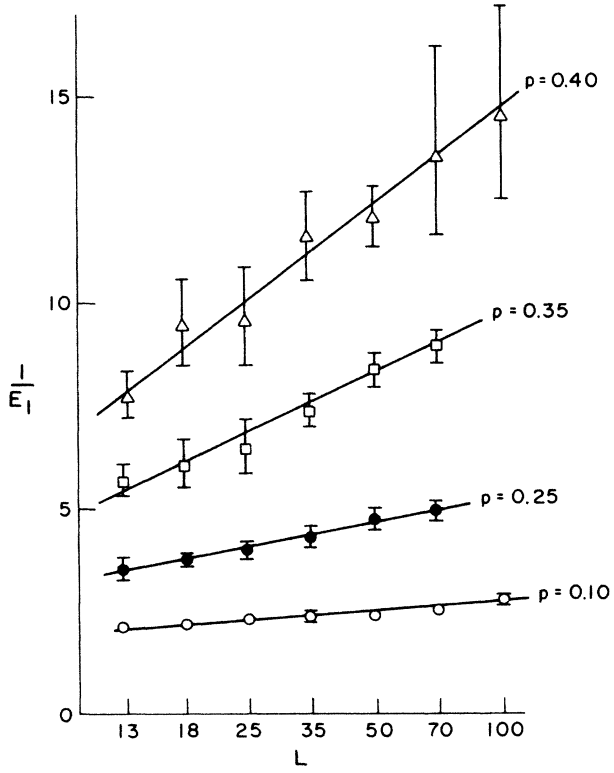

FIG. 4. The inverse of the breakdown field vs the system size L on a logarithmic scale. The linear dependence demonstrates that E_1 has the form given by Eq. (6). The lines are fits to Eq. (6).

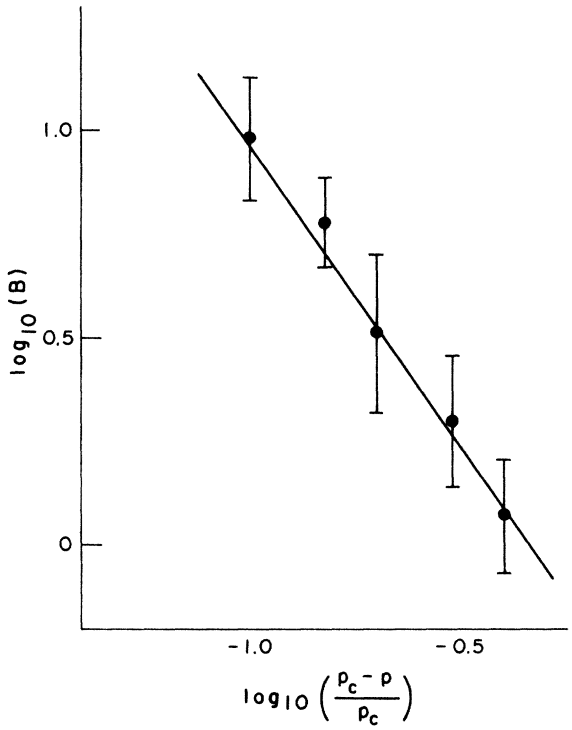

FIG. 5. This is a plot of $\log_{10}[B(p)]$ vs $\log_{10}[(p_c-p)/p_c]$ to test the relation $B(p)\approx(p_c-p)^{-\nu}$. The slope of the line is predicted in Eq. (3) to be $-\nu$ where ν is the percolation correlation length exponent. The power-law fit to the data gives $\nu=1.46\pm0.22$ which is consistent with the two-dimensional value $\nu=\frac{4}{3}$.

will give an outline of the derivation of the breakdown field distribution function and show that the result is consistent with the numerical distribution function found from the computer simulations. The details of this derivation are given in Refs. 5 and 8. The breakdown field is determined by the linear size, $l_{\max}$, of the largest defect in the sample.[2,3] Let $C_L(l_{\max})$ be the probability that no defect larger than size $l_{\max}$ exists in a d-dimensional hypercubical volume L^d. We then subdivide the cube with volume L^d into $(L/L_1)^d$ smaller cubes of linear dimension L_1. If the characteristic size of the largest defects is much smaller than L_1, then, by using the statistical independence of the subcubes, we arrive at the result that the probability of there being no defect larger than $l_{\max}$ on the L^d lattice is

$$[C_{L_1}(l_{\max})]^{(L/L_1)^d}\approx C_L(l_{\max}) . \tag{7}$$

By solving this equation, along with the fact that the percolation cluster size is an exponentially decaying function of l,[9] we arrive at the result

$$C_L(l_{\max})=\exp[-cL^d\exp(-kl_{\max})] . \tag{8}$$

This is the form first derived by Duxbury and Leath[5,8] and is the appropriate distribution function far from the percolation threshold, i.e., if $L\gg\xi$. If the percolation cluster size distribution is an algebraically decaying function of n then the distribution function is

$$C_L(l_{\max})=\exp(-cL^d l_{\max}^{-m}) . \tag{9}$$

This distribution is of the Weibull form and is the appropriate distribution close to the percolation threshold, i.e., when $L\ll\xi$.

We can use this result to give the distribution function for the dielectric breakdown field strength in this model. We argued earlier that that enhancement of the electric field in the neighborhood of a linear defect is proportional to the linear dimension of the defect. The breakdown field is of order $1/l_{\max}$ where $l_{\max}$ is the size of the critical defect so the breakdown field distribution function $F_L(E)$ is of the form

$$F_L(E)\approx 1-\exp\left[-cL^d\exp\left[-\frac{k}{E}\right]\right] . \tag{10}$$

This form is valid if $L\gg\xi$. The function $F_L(E)$ is the probability that a sample of size L will experience a dielectric breakdown failure if an external electric field E is applied to the sample. For $E\to 0$ the probability of failure tends to zero and as $E\to\infty$ the probability of failure tends to unity. The constant k is proportional to the inverse of the percolation correlation length and c is weakly dependent on p.[5,8] The connection of this analysis to the logarithmic scaling of the average breakdown field can now be made clear. The average breakdown field will be close to the value $E_{1/2}$ which is defined as the value of E for which half of the samples will fail.

$$\tfrac{1}{2}=F_L(E_{1/2})=1-\exp\left[-cL^d\exp\left[-\frac{k}{E_{1/2}}\right]\right] . \tag{11}$$

This can be easily solved for $E_{1/2}$:

$$E_{1/2}=\frac{1}{A(p)+B(p)\ln(L)} , \tag{12}$$

where

$$A(p)=\frac{\ln(c)-\ln[\ln(2)]}{k} , \tag{13}$$

and

$$B(p)=\frac{d}{k} . \tag{14}$$

Both $A(p)$ and $B(p)$ should scale proportional to the correlation length as p approaches the percolation threshold. This is exactly the form presented above [Eq. (6)].

The equivalent distribution for the Weibull form is

$$F_L(E)=1-\exp(-cL^dE^m) , \tag{15}$$

and the average breakdown field scales as

$$E_{1/2}\approx L^{-d/m} . \tag{16}$$

We will now demonstrate that the distribution form (10) gives a much better fit to the breakdown distribution

function numerical data than the Weibull form (15) if p is far from the percolation threshold. In Fig. 6(a) we plot the logarithm of the logarithm of the distribution function versus the inverse of the breakdown electric field. In Fig. 6(b) we plot the same function versus the logarithm of the breakdown electric field. If Eq. (10) is appropriate, the data for the two different lattice sizes should collapse onto a single straight line in Fig. 6(a). If the Weibull form (15) is appropriate then the data will collapse to a single straight line in Fig. 6(b). As can be seen, the data form a very nice straight line in Fig. 6(a) whereas the data in the Weibull plot [Fig. 6(b)] are noticeably curved. The slope of the curve in the Weibull plot [Fig. 6(b)] is the Weibull exponent m. The data indicate that m is quite large ($m \approx 20$). This is a good empirical signal that (10) may be the more appropriate distribution function to use.

We have also performed a few simulations of three-dimensional metal-loaded dielectrics to make sure that

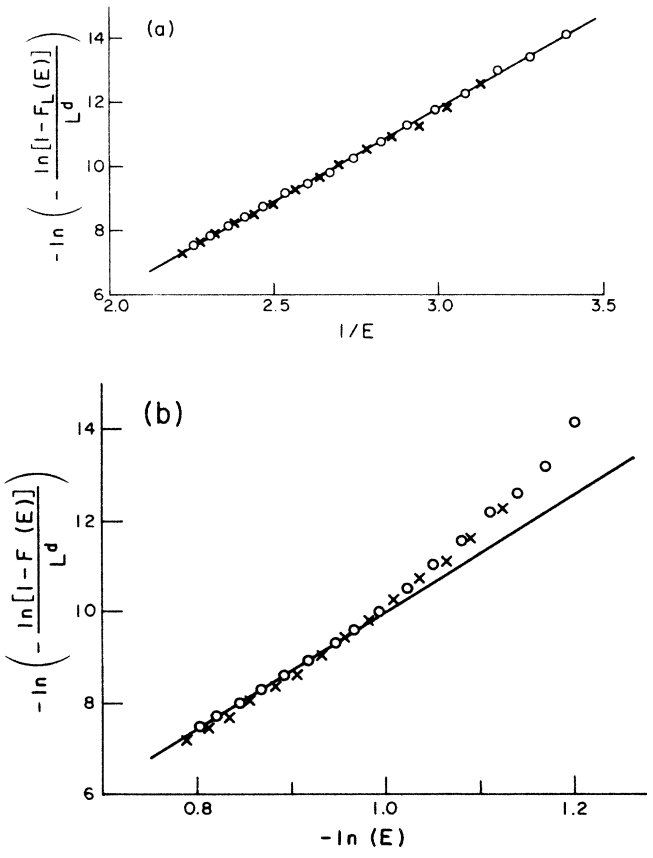

FIG. 6. (a) Data for the breakdown field distribution function plotted in a form to test the Duxbury and Leath form [Eq. (10)] for the distribution function. If this form is valid, the data for the two different lattice sizes should collapse to a single straight line. (b) The same data plotted in a form to test the Weibull form [Eq. (15)] for the distribution function. The straight line is a guide to the eye and has a slope (Weibull exponent) of order $m = 20$. The Duxbury and Leath form (10) for the distribution function gives a much better fit to the data than the Weibull form.

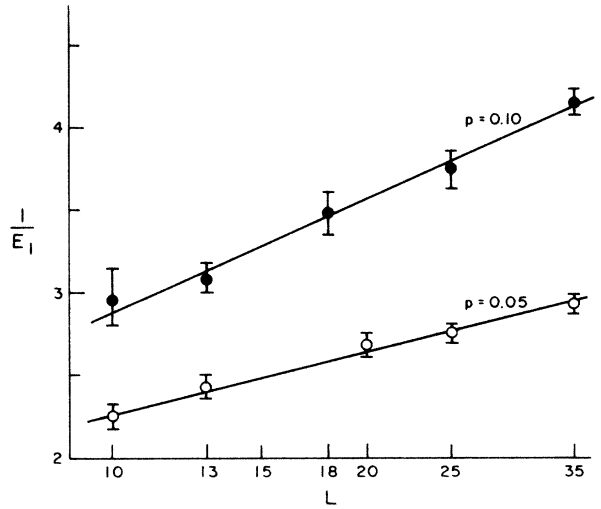

FIG. 7. The inverse of the initial breakdown field vs the logarithm of the system size in a three-dimensional model for $p = 0.100$ and 0.050. The percolation threshold for this model is $p_c \cong 0.25$. The linear behavior of the data in this plot confirms that E_1 is of the form (6) in three dimensions and indirectly supports the form (10) for the breakdown distribution function in three dimensions. The lines in the figure are fits to Eq. (6).

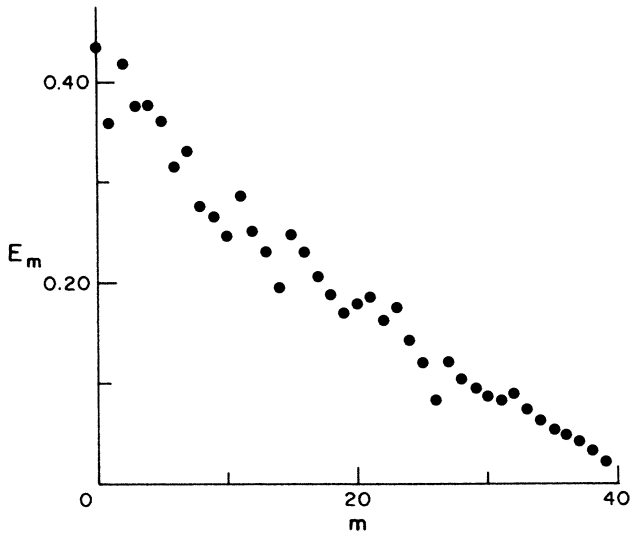

FIG. 8. The breakdown field, E_n, in a two-dimensional sample of size $L = 48$ with $p = 0.100$ (the sample shown in Fig. 1) as a function of the number of prior microscopic dielectric breakdowns, n. Note that E_n is roughly a decreasing function of the number of previous breakdowns. The complete breakdown field is the largest of the microscopic breakdown fields and the initial breakdown field is the first point. In this particular case, the initial and complete breakdown fields are the same. In general, the complete breakdown field is larger than the initial breakdown field but the average initial breakdown field and the average complete breakdown field turn out to be the same to within the the natural distribution width of the two quantities.

our arguments extend properly to higher dimensions. We expect the critical defect to be a roughly linear object as shown in Fig. 2 in all dimensions. Therefore the arguments we presented for the breakdown field distribution function should also be valid in three dimensions. To save computer time, instead of determining the entire distribution function for three-dimensional breakdown, we determined the average breakdown field at several metal fractions, p, and several sample sizes L. If the distribution function of breakdown fields is of the form given by Eq. (10) then the average breakdown field should have the size dependence given by Eq. (6). A plot of the data demonstrating this is shown in Fig. 7. The linear dependence of $1/E_1$ on $\ln(L)$ confirms our predictions.

Finally, we would like to mention the connection of the initial breakdown field to the complete breakdown field. The crucial question to be answered is, does initial failure cause the system to undergo a catastrophic sequence of microscopic failures resulting in the formation of a macroscopic breakdown path? We can answer this question by continuing our simulations as follows. After each failure we replace the failed capacitor by a conductor and re-solve for the equilibrium voltage distribution. We continue this process until a conducting path is formed across the system. We define the complete breakdown field as the largest breakdown field found in that sequence of microscopic failures. A plot of the breakdown field as a function of the number of previous microscopic failures is given in Fig. 8. On average the function is a decreasing function of the number of previous failures. This is because the critical defect is getting longer and longer as the breakdown process continues so the breakdown field gets smaller. The complete breakdown field was typically slightly larger than the initial breakdown field but in every case we have tested the uncertainty of the complete breakdown field strongly overlaps the uncertainty in the initial breakdown field. We conclude that the initial and complete breakdown fields have essentially the same value.

IV. CONCLUSIONS

Based on the theoretical arguments and the numerical data presented in this paper we can suggest a couple of crucial experiments for dielectric breakdown in metal-loaded dielectrics. First, the initial dielectric breakdown field should be measured in a large number of similarly prepared random mixtures of metal and dielectric. This should be done as a function of metal fraction and system size. The resulting distribution functions can be plotted in the manner given in Fig. 6(a) to test the Duxbury and Leath form (10) for the breakdown distribution function. The manner in which the average breakdown field depends on p and L can be tested against the expected form (6). One can test whether the coefficient $B(p)$ is proportional to the correlation length in the sample. Finally, a careful study can be made of the difference between the initial and complete breakdown fields. We predict that they are essentially the same.

ACKNOWLEDGMENTS

We wish to thank Paul Leath, Eugene Stanley, and Mike Thorpe, and David Srolovitz, Julia Yeomans, David Bowman, Josephine Covino, Phil Gibson, and Ron Larson, and Rod Perala at Electromagnetic Applications in Denver for useful conversations. We also thank the National Science Foundation for grants of supercomputer time at Colorado State University, and the John von Neumann Computer Center at Princeton.

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[11]This calculation can be done exactly for a pair of infinitesimally thin metallic needles oriented along the applied field. If the length of the needles is l and the distance between the two defects is much less than l, the field enhancement between the two clusters is of order $l/\ln(l)$. The breakdown field will be reduced by the same factor. The logarithmic correction is difficult to observe in a random network. A single defect, by comparison, has a field enhancement at the tip of order $l^{1/2}/\ln(l)$.

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