Electrical Breakdown Measurements of Semicontinuous Metal Films

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The breakdown current I_c of a percolating system is measured on real materials. We find $I_c \propto B^{-x}$; B is the weakly nonlinear response determined by third-harmonic generation. A new criterion for I_c is suggested, defined as the current at which a hot spot reaches the melting temperature of the metallic grains, T_m. This criterion remains valid in the presence of nonlinear effects. It is also consistent with the experimental observations: At I_c the resistance either increases to infinity or decreases, i.e., the local geometry is changed at T_m. Modeling the breakdown by hot spots yields the above power law with $0.5 \geq x \geq 0.36$, in excellent agreement with the measured data: $x = 0.48$ and 0.41 for Ag and Au films.

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The breakdown behavior of inhomogeneous or composite materials is of great fundamental as well as practical interest. The physical properties of composites are usually characterized by universal power laws, where the critical exponents depend only on the dimensionality. However, some properties show highly nonuniversal behavior, where the critical exponents are extremely sensitive to fine details of the microgeometry ($1/f$ noise for example). This is true in the case of breakdown processes, such as mechanical, dielectric, or electrical breakdown. In random (or disordered) superconductor-dielectric mixtures [1,2], the critical current I_c is found to vanish above the metallic percolation threshold p_c as $I_c \propto (p-p_c)^y$, where p is the volume fraction (probability) of the superconductor. Since the sheet resistance R follows a different power law, $R \propto (p-p_c)^{-t}$, with a fairly universal exponent $t_{2D} = 1.3$, $I_c \propto R^{y/t}$. Estimation of y by the nodes, links, and blobs (NLB) model [1,2] yields $y = \nu$ (in two dimensions), where ν is the critical exponent of the percolation correlation length ξ ($\nu_{2D} = \frac{4}{3}$). This prediction is in agreement with experiments on superconductor-dielectric mixtures [1]. Computer simulations on the random fuse model, where each bond is either an insulator or a fuse (with a certain burning current i_c, above which it becomes insulating), yielded $y = 0.82 \pm 0.1$ [3]. Within the numerical uncertainty, this result agrees with the calculation below the percolation threshold: The critical electric field E_c follows a power law with $y = 1.1 \pm 0.2$ [4], and from the duality theorem I_c (the breakdown current) should vanish with the same critical exponent (in 2D). Higher values of y are expected in the case of continuum percolation [2]. It should be noted that the breakdown of an actual realization of a percolative network depends essentially on the *weakest link*, not the average one, leading to a lower breakdown current than the NLB estimation. This is in contrast with the superconductor-dielectric case where the current is distributed according to the number of links regardless of their strength. Moreover, the probability of an extremely weak link increases with system size, and the breakdown current should vanish logarithmically with its linear size L [5]. The above arguments lead to $y \geq \nu$, in contrast with the finite-size numerical simulations quoted above [3,4]. In particular, the breakdown voltage $V_c \propto (p-p_c)^{y-t}$ vanishes at p_c if $y > t$ and diverges if $y < t$. As the NLB model predicts that V_c vanishes at p_c, we conclude that it should also vanish in the more extreme case, and hence y should be larger than t.

We report here, for the first time, an actual measurement of the breakdown current I_c in real materials: semicontinuous Ag and Au films. We find that y is appreciably larger than ν for both Ag and Au films, and is not the preferred exponent for characterizing the breakdown. We find that a better characterization is provided by the scaling relation $I_c \propto B^{-x}$, where B is the normalized third harmonic generated by the film ($B = V_{3f}/I^3$), resulting from local Joule heating [6,7]. Both the breakdown current I_c and the third-harmonic coefficient B are modeled by a hot-spot description, where B measures the local temperature rise at the hot spots. Breakdown occurs whenever a hot spot reaches the melting temperature of the metallic grains, T_m. In that case an irreversible change occurs and the film resistance is modified. The resulting resistance may be either larger or smaller than the original value. In the first process, a weak link is disconnected, like a fuse, while in the latter case a very thin channel becomes wider, resulting in a decrease of the film resistance. In both cases the local power dissipation is reduced (the experimental setup regulates the current up to a certain voltage limit). The description is somewhat different, and more general, than the usual approach to breakdown phenomena because (1) the critical exponent x is almost insensitive to the microgeometry and (2) the above power law holds even in the regime where hopping conductivity is not negligible, in contrast to the scaling of I_c versus the film resistance R.

We have measured the breakdown current and the third-harmonic generation of thin semicontinuous Ag and

Au percolating films. The films were evaporated under a vacuum of 10^{-6} torr at a rate of 0.1 nm/sec onto room-temperature glass substrates. Several samples with different values of surface coverage were prepared simultaneously in each run. The size of each sample was 4 by 1 mm^2. The samples were then removed from the vacuum system and measured at room temperature. Measurements of the third harmonic generated by the samples were done using a resistive balance bridge connected directly to an HP35660A dynamic signal analyzer, as described elsewhere [7]. The *I-V* characteristic was measured by the four-probe method, using a dc current source with a programmable voltage limit. At low currents the film shows Ohmic behavior (R constant). At higher currents, the resistance increases due to Joule heating, i.e., $dR/dI > 0$. When the current is further increased, dR/dI exhibits several discontinuities and finally the sample becomes insulating or undergoes a significant resistance change (see Fig. 1). The breakdown current I_c was defined as the current at which the first irreversible change was measured, i.e., the first discontinuity in dR/dI, and not as the final breakdown achieved at yet higher currents. This final breakdown process depends upon the details of the breakdown of the first links and we find that it does not obey any simple scaling law. During the later stage of the breakdown process several samples suffered rapid resistance changes: Under constant current and fixed voltage limit their resistance showed large fluctuations in time, between finite and infinite resistance states. A detailed description of the phenomena that occur during the approach to the final breakdown will be published separately.

The scaling of I_c as a function of the film resistance R is shown in Fig. 2. The measurements provide a wide span of resistances and critical currents: 3 decades of R and 4 decades of I_c. For the Ag films $I_c \propto R^{-a}$, $a = 1.75 \pm 0.3$, which is higher than the numerical-simulation value $a = y/t \simeq 0.85$ for lattice percolation [3,4]. In particular, $y > \nu$ as expected for both the large system size ($L \gg \xi$) and its continuum nature. For the Au films a linear fit does not seem to describe the data properly. (A single straight line has systematic deviations; the experimental points are *above* such a line at low and high resistances and *below* it at the intermediate zone.) Instead, one can identify two critical exponents: $a = 1.75 \pm 0.4$ for $R < 2$ kΩ and $a = 0.85 \pm 0.2$ for $R > 2$ kΩ. Separating the Au samples into two regimes, according to the film resistance, is consistent with the behavior of the third harmonic [7] which is exhibited in the inset of Fig. 3(b). For the high-resistance Au films, a is consistent with the numerical-simulation value for the random fuse model. This result is misleading, however, as in these films a hopping conductivity at the hot spots is initiated by the local temperature rise (see Ref. [7]). Such an effect is not included in the random fuse model; hence we take this agreement to be a coincidence. Plotting the critical current as a function of the third-harmonic coefficient B, rather than R, yields $I_c \propto B^{-x}$ for both the Ag and the Au films, as shown in Fig. 3. Note that for the Au films, a single power law describes the entire range of film resistances, unlike the plot of I_c vs R.

The experimental data discussed above show a clear correlation between the third-harmonic coefficient B and the breakdown current I_c. The physical origin of the third harmonic is known to be the local temperature in-

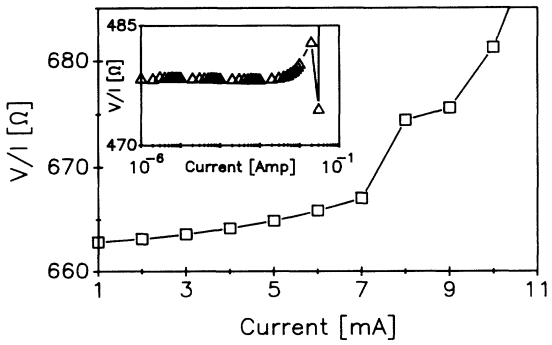

FIG. 1. The breakdown process of one of the Au films. Discontinuities in dR/dI appear at 8 and 10 mA, while the film becomes insulating at 20 mA. The breakdown current in this case occurs at 8 mA. Inset: A similar picture for an Ag film (logarithmic current scale). In this case the resistance is *reduced* at 30 mA and the film becomes disconnected at 40 mA.

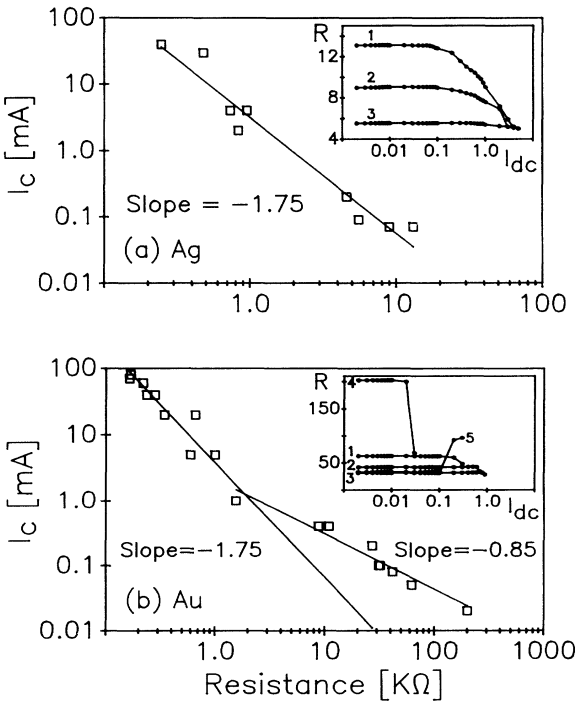

FIG. 2. Scaling of I_c as a function of the film resistance R. Inset: Several breakdown cycles of a high-resistance film (resistance in kΩ and current in mA). (a) Ag films. (b) Au films.

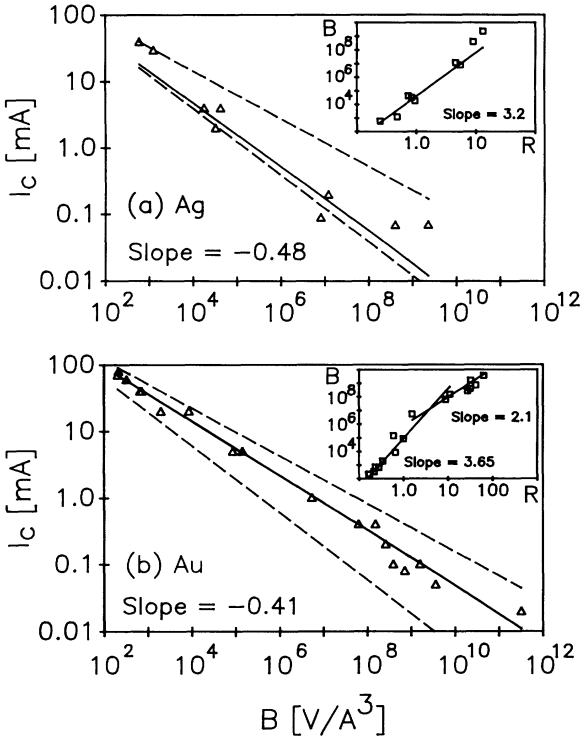

FIG. 3. Scaling of I_c as a function of the normalized third-harmonic coefficient B. The straight lines describe the power law $I_c \propto B^{-x}$, with $x=0.48$ and $x=0.41$ for the Ag and the Au films, respectively. Dashed lines are for the lower and upper bounds of x (0.36 and 0.5). Inset: The scaling of B with R (in kΩ), where the corresponding critical exponents are taken from Ref. [7]. (a) Ag films. (b) Au films.

crease [6]; we propose that local heating is also at the origin of an irreversible geometrical change in the case of I_c. Assuming that this change is due to local melting, we now show that I_c scales as B^{-x}, where $0.36 \le x \le 0.5$. For clarity we first derive B in terms of the local temperature rise ΔT and then calculate the breakdown current I_c. The temperature rise due to a weak link with resistance r_0 and current i is $\Delta T = i^2 r_0 h_{HS}$, where h_{HS} is the ratio between the temperature rise and the power dissipated in the hot spot. The local resistance change is given by $\delta r = r_0 \beta \Delta T$, where $\beta = (1/r)dr/dT$. Applying an ac current $I = I_0 \cos(\omega t)$ results in the generation of a third-harmonic voltage component $V_{3f} = (1/4I_0)\sum i^2 \delta r$, where the sum is over all the hot spots. Assuming that both r_0 and h_{HS} are the same for all links, the normalized third-harmonic component $B = V_{3f}/I_0^3$ is given by $B = (1/4I_0^4)\beta h_{HS} r_0^2 \sum i^4$. For an $L \times L$ resistor network the current in each resistor is I/L (L is measured in units of the lattice constant); hence

$$B = \tfrac{1}{4} \beta h_{HS} r_0^2 / L^2 \qquad (1)$$

and the temperature change is $\Delta T = \frac{1}{2} r_0 h_{HS} (I_0/L)^2 [1 + \cos(2\omega t)]$. For a diluted network the current in some resistors is much larger and the third harmonic scales as $(p - p_c)^{-(2t+\kappa)}$, where κ is defined through $\sum i^4 / (\sum i^2)^2 \propto (p - p_c)^{-\kappa}$. A lower bound for B (and κ) is obtained by taking into account only the singly connected (SC) bonds [8]. For a large two-dimensional system of linear size $L \gg \xi$, the number of SC bonds is $N_{SC} = (L/\xi)^2 \times (p - p_c)^{-1}$ (ignoring a constant of order unity) and the average current through each SC bond is $I_{SC} = (\xi/L) I_0$; hence B is given by

$$B \ge (1/4I_0^4) \beta h_{HS} r_0^2 N_{SC} I_{SC}^4 . \qquad (2)$$

The average ac component of the temperature rise in each of the SC bonds is $\Delta T_{SC} = \frac{1}{2} h_{HS} r_0 I_{SC}^2$; thus

$$B \ge (1/2I_0^2) \Delta T_{SC} \beta r_0 (p - p_c)^{-1} . \qquad (3)$$

Let ΔT_m be the temperature rise required to reach the melting temperature of the metallic grains. The breakdown current I_c may be defined as the current at which the melting temperature is reached:

$$I_c \ge (\Delta T_m \beta r_0 / 2)^{1/2} (p - p_c)^{-1/2} B^{-1/2} . \qquad (4)$$

Using $B_0 = \frac{1}{4} \beta h_{HS} r_0^2 / L^2$, $I_{c0} = L (2 \Delta T_m / r_0 h_{HS})^{1/2}$, which are the values of B, I_c appropriate for an undiluted uniform network, and noting that $(p - p_c)^{-1/2} = (B/B_0)^{1/2(2t+\kappa)}$, one finds

$$I_c \ge I_{c0} (B/B_0)^{1/2(2t+\kappa) - 1/2} . \qquad (5)$$

Substituting the lower bound of κ ($\kappa = 2\nu + 1 - 2t$, as calculated by the singly connected bonds [8]) yields $I_c \propto (p - p_c)^{\nu}$ in agreement with the NLB calculation [1,2]. In the case of continuum percolation, κ may be much larger due to the high current densities in some very thin channels. Typically, only a fraction of the singly connected bonds will control the sum $\sum i^4$; hence I_c is expected to scale as B^{-x} where $\frac{1}{2} \ge x \ge \frac{1}{2} - 1/2(2t + \kappa)$. The upper bound is for the case where a single hot spot controls both B and I_c. In two dimensions $t \simeq 1.3$ and $\kappa \simeq 1.1$ [9]; hence $0.36 \le x \le 0.5$. Substituting $\Delta T_m \simeq 10^3$ K, $r_0 = 1$ Ω, and $\beta \simeq 10^{-3}$ K^{-1} [7] yields $\Delta T_m \beta r_0 / 2 \simeq 0.5$; hence $I_c = I_{c0} (B/B_0)^{-x}$ with $I_{c0} B_0^x \simeq 0.7$ (the prefactor is not sensitive to the exact value of x), in excellent agreement with the measured power law for both the Ag ($x = 0.48 \pm 0.05$, $I_{c0} B_0^x = 0.4$) and the Au ($x = 0.41 \pm 0.01$, $I_{c0} B_0^x = 0.6$) samples (see Fig. 3). Moreover, all the experimental points (except for one Ag film) are bounded by the lower and upper values of x, i.e., $I_{c0} (B/B_0)^{-0.36} \ge I_c \ge I_{c0} (B/B_0)^{-0.5}$.

In the case of high-resistance Au films ($R > 10$ kΩ), it is shown in Ref. [7] that weak links are short circuited by a hopping process initiated by the temperature rise at the hot spots. The current densities at the weak links are thus decreased and the breakdown current increases [Fig. 2(b)]. This effect is detected by third-harmonic measurements, where B increases more slowly with increasing resistance at the high-resistance regime [see the inset of

Fig. 3(b)]. We take the validity of (5) for these high-resistance Au films as an experimental verification to our approach for the breakdown process.

We turn now to a brief discussion of the possible microscopic processes involved in the breakdown of such films. In the case of the low-resistance samples, the breakdown usually resulted in an insulating film; thus *all* the links that carried high current densities were burned. Applying a high voltage, protected by a very low current limit, caused the sample to be reconnected. The resulting film had a high resistance and an extremely low breakdown current, showing that only *a few* links were established. The new film is now dominated by red links (singly connected links) and the backbone statistics is totally different. We call these films "unstable." The breakdown of the high-resistance films ($R > 10\ \mathrm{k}\Omega$) resulted in higher, lower, or infinite resistance, as shown in the inset of Fig. 2(b), where several breakdown cycles of the same sample are shown. This picture means that a few red bonds were either burned out (fuselike), established (dielectric breakdown), or improved (channel width increase). The resistance of the "new" film, after each breakdown cycle, was always larger than $10\ \mathrm{k}\Omega$, and its breakdown current and third-harmonic coefficient were consistent with the new resistance. This picture indicates that the electrical current path was changed after each breakdown process while the backbone statistics was unchanged (i.e., the topologies before and after the breakdown are the same). We call these films "stable." A high-resistance film could suffer several "stable" breakdown cycles until it became "unstable" in the sense mentioned above. The so-called stable and unstable breakdown processes are thus dependent on the statistics of the new backbone.

Another interesting aspect of the microscopic breakdown process is the decrease in the film resistance, which was the normal behavior of the high-resistance Ag films [see the inset of Fig. 2(a)]. Such a behavior seems to justify the hot-spot picture and the melting-temperature criterion. Whenever a hot spot reaches the melting temperature, the actual geometrical change depends mainly on surface tension effects: A weak link may be either disconnected (like a fuse) or improved (become wider). In both cases the local power dissipation is reduced (in the latter case the current is kept constant). Considering such effects, preparation of films with uniform properties is possible using a training process, i.e., eliminating weak links by successive breakdown processes.

In summary, we have measured the breakdown current I_c of thin semicontinuous Ag and Au films and found that I_c vanishes as B^{-x}, with $x = 0.48 \pm 0.05$ and 0.41 ± 0.01 for the Ag and Au films, respectively. Modeling the breakdown process in terms of hot spots yields such a behavior with $0.36 \lesssim x \leq 0.5$. Excellent agreement is found between the calculated and measured data, not only for the critical exponent x but also for the prefactor $I_{c0}B_0^x$. In this approach, the breakdown current is defined as the current at which a hot spot reaches the melting temperature of the metallic grains, in contrast to the usual definition (as in the fuse model): The critical exponent x is almost insensitive to fine details of the microgeometry while y is extremely sensitive to such details, and the power law $I_c \propto B^{-x}$ holds in the presence of nonlinear effects (like the hopping process) unlike the scaling of I_c with $p - p_c$. The breakdown current, within this description, is sensitive to several physical conditions which are absent in the usual breakdown picture: (1) the existence of extremely narrow necks carrying a high current density, (2) interaction between hot spots, resulting in a higher local temperature, (3) excitation of other local conductance processes such as hopping due to the high temperature at the hot spot, and (4) the heat transfer efficiency of the coolant and the substrate, i.e., the critical current I_c of a film depends on the external conditions. We conclude that measurements of third-harmonic generation can be used as a tool for nondestructive study and prediction of breakdown. Higher moments of the current distribution (determined by higher harmonics) should be even more sensitive to crucial details of the microgeometry and could thus be an even more sensitive tool, as discussed by Bergman [10]. This technique is not limited to electrical measurements, and can be applied to mechanical properties as well.

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