Critical Behavior of Thermal Relaxation near a Breakdown Point

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At a composition far above the percolation threshold, the resistance of a composite sample increases with time due to Joule heating as a constant current of a sufficiently large value is passed through the sample. If the current is less than a certain breakdown current (I_b) the resistance eventually reaches a steady value with a characteristic relaxation time τ_h. The latter diverges with current I as $\tau_h \sim (1 - I^2/I_b^2)^{-z}$. The value of the exponent z displays large fluctuations leading to unusual scaling of the relaxation time. It is shown that the results lead to important conclusions about the nature of breakdown phenomena.

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Breakdowns or fractures in random systems form an important class of nonthermodynamic phase transitions [1]. Much of the effort [1–9] to understand such irreversible phenomena naturally uses the framework already developed for thermodynamic critical phenomena although descriptions remain far from complete. Consider, for example, the order of a breakdown transition. Zapperi *et al.* [5] have suggested that the latter is first order as a function of the external field. The relevant "order parameter," elastic constant, or conductivity suffers a discontinuous change from a finite value to zero at a breakdown point. Using a somewhat different model, Andersen *et al.* [6] supported the idea of a first-order transition but only at small disorder. They predicted a change to a second-order transition at higher disorder, thus indicating the presence of a tricritical point. On the experimental side, some recent results on electrical failure in composites suggest rather conflicting scenerios. When a sufficiently large current I is passed through a composite sample (a random binary mixture of a conductor and an insulator [10]) with the conducting fraction p typically far above the percolation threshold $p_c \ll p$, the Joule heating causes the sample resistance to increase. For currents greater than the breakdown current I_b, heating eventually leads to an irreversible electric breakdown. It was found [8] that when a constant current $I > I_b$ is passed the resistance of such a sample increases as a power of time, $R \sim [\tau_h(I) - t]^{-0.65}$. The relaxation time (or time to failure) τ_h also exhibits a critical behavior (see below). However, measurements [9] starting with small currents show that the sample conductance drops to zero as soon as the current equals or exceeds the breakdown current, thereby signaling a first-order transition.

Clearly, it will be useful to obtain further information on the transition. In this Letter, we report the results of our dynamic measurements of thermal relaxation using currents *less* than, but up to, the breakdown current in composite samples with *varying* amounts of disorder. The observed relaxation behavior is reminiscent of the well-known phenomena of slowing down [11] near critical points in thermodynamic systems. The relaxation times also exhibit an unusual scaling relation due to a strong dependence on disorder. The present results together with the earlier ones [8] constitute, to our knowledge, the first full description of a dynamic breakdown phenomenon *both* above and below the critical parameter. We examine below various features including the interplay of disorder and breakdown dynamics, which becomes significant particularly in the regime of weak disorder $p_c \ll p \leq 1$. Few related experimental works include investigation of avalanche dynamics [12] and strain-relaxation measurements in metal networks near the percolation threshold [13].

Measurements were performed in composites of carbon and wax (C-W) under a constant dc current condition at room temperature and the sample resistance was monitored as a function of time during heating. The preparation and characterization of the system have been described earlier [14]. p_c in the C-W system is 0.76% (by volume). In the tunneling regime [15] close to p_c, the resistance *decreases* with bias. It has been recently found out that there exists a Joule regime in, or a conducting fraction p_J above, which the resistance always *increases* with bias [9]. The nominal carbon fractions of the samples ranging from 4.5% to 10% were above or near p_J which is approximately 4.5% [16]. Samples prepared with larger carbon fractions were mechanically unstable. On the other hand, samples with lower carbon fractions had a tunneling effect offsetting the effect of Joule heating (see Fig. 1 of Ref. [9]). The geometry of current flow in a cylinderical sample is illustrated in the inset in Fig. 1. Various properties of the samples used in the present work are given in Table I. In a typical measurement, the current corresponding to a certain value I would be turned on at time $t = 0$ and the bias across the sample would be measured at an interval of time (usually 250 msec) until it reached a steady value when a balance between dissipation and generation of heat within the sample is established. The sample was then allowed to cool for up to 2 h before measurements would be repeated with an another current, not exceeding the breakdown current. The relaxation during cooling from a hot steady state was also

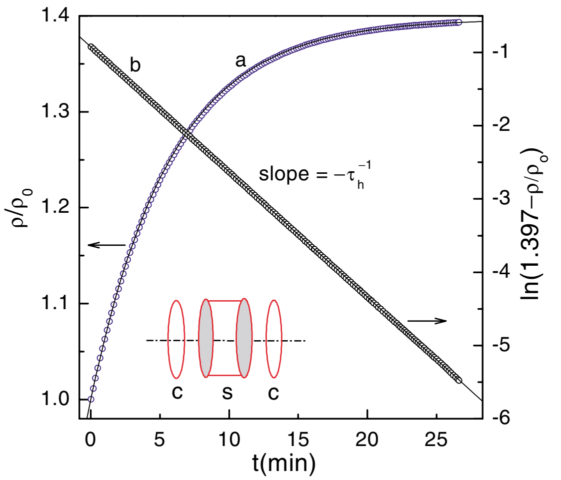

FIG. 1 (color online). A typical resistance relaxation curve (*a*) of a cylindrical sample (No. 3) heated by passing a constant current of 68.5 mA. Many data points have been omitted for clarity. The solid line is a fit to a simple exponential $\rho/\rho_o = 1.397 - 0.397\exp(-t/\tau_h)$. The same data are also shown in log-linear plot (*b*). The slope of the straight line yields the negative inverse of the relaxation time $\tau_h = 348$ s. The inset is an assembly diagram showing copper foils (c) used as electrodes and attached to the sample (s) with silver paints.

measured. It exhibited an anamalous behavior. This will be reported elsewhere.

Figure 1 shows a typical relaxation curve (*a*) during heating, obtained from a cylinderical sample (No. 3). The curve is well described by a simple exponential process indicated by the solid line in Fig. 1. It may be recalled [17] that the relaxation function $f(t)$ is almost universally given by $f(t) \sim \exp[-(t/\tau)^\alpha]$. Here t is time, τ is a relaxation time constant, and α is an exponent. The exponential relaxation corresponding to $\alpha = 1$ occurs mostly in simple systems such as homogeneous ordered solids [18]. A simple exponential in the present case may be attributed to a long-time relaxation behavior. In systems with characteristic time scales such as percolating networks, the relaxation function may change from a stretched exponential at short times to a simple exponential at long times [13,19]. For samples with large $p \gg p_c$ such as the ones used here, the crossover times should be quite small. Close to p_c, α was determined to be about 0.8 [13] in elastic relaxation in dilute metal foils, and about 0.4 [20] in voltage relaxation in the same system as the present one.

TABLE I. Various properties of the samples used. p is the carbon fraction by volume (%). ρ_o and τ_{ho} are the resistivity and relaxation time at zero current. I_b is the breakdown current and z is the exponent in Eq. (1). Each number in the last row is the linear-correlation coefficient of the respective column variable and z.

Sample No.	p[a]	Height (mm)	ρ_o (Ω cm)	τ_{ho} (sec)	I_b (mA)	10^3 z
1	7.5	6.0	567	495	31.5	175 ± 7
2	4.5	4.1	561	145	19.2	187 ± 5
3	7.5	4.1	280	168	73	300 ± 14
4	4.5	2.7	214	455	83.1	76 ± 3
5	7.5	2.8	191	113	112	180 ± 8
6	7.5	4.2	188	156	95	150 ± 11
7	7.5	$10 \times 4 \times 2$[b]	15.2	67	18.7	230 ± 7
8	10	8.3	5.26	194	135	173 ± 8
			−0.05	−0.56	−0.22	

[a] Nominal value.
[b] This sample is ribbon shaped but all other samples are cylinders of 10 mm diameter.

The relaxation time in several samples of different compositions and dimensions (see Table I) against current are shown in Fig. 2. τ_h of a sample was found to diverge as a power law

$$\tau_h = \tau_{ho} \mid \epsilon \mid^{-z}. \tag{1}$$

Here, τ_{ho} is a prefactor, $\epsilon = (I^2/I_b^2 - 1) < 0$, and z is a dynamical exponent. τ_{ho}, I_b, and z were treated as fitting parameters and its values are listed in Table I. τ_{ho} is really the relaxation time at zero current. It depends on sample dimensions [see Eq. (2)] but, more importantly, monotonically increases with disorder. In fact, τ_{ho} diverges as p approaches p_c. Such disorder-induced divergence of relaxation time has been qualitatively observed by Ghosh *et al.* [13]. The breakdown current I_b scales with R_o as $I_b \sim R_o^{-0.44}$ [9] with a high degree of scatter. R_o is the resistance at zero current.

The exponent z thus determined was not a constant even within experimental uncertainties of 5%. It exhibits an unusual sample-to-sample fluctuation as seen in Fig. 3 where it is plotted against a quantity r as a measure of disorder. (It is not practical to use p due to uncertainties in its values.) r is defined as the ratio of the zero-current

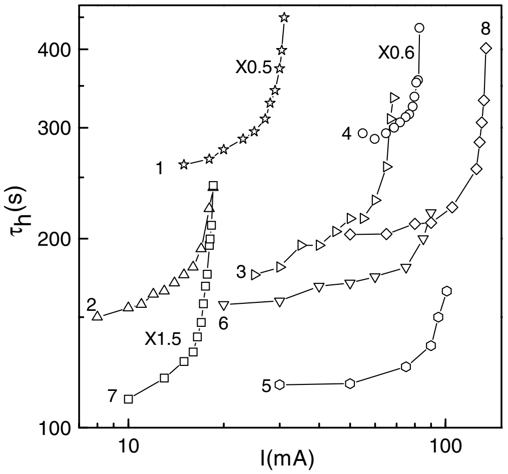

FIG. 2. Relaxation time vs current in various samples as in Table I. Some data have been translated vertically by factors as marked.

sample resistivity and the resistivity of the pure conducting phase ($\sim 10^{-2}$ Ω cm). r is 1 at zero disorder ($p = 1$) and increases monotonically with disorder as p decreases from 1. Within the range of disorder (2 orders of magnitude) investigated in this work, the exponent has an average value of 0.184 ± 0.06, i.e., 33% uncertainties. The highest value (0.3) is about 4 times the lowest one (0.076). Without these two extreme values, uncertainties reduce to 17% which is still beyond the experimental error. Clearly, the relaxation exponent z for $I < I_b$ is disorder dependent. In fact, one can classify the breakdown-related exponents, both theoretical and experimental, into two groups: (i) disorder dependent and (ii) disorder independent or "universal." Examples of the first group include, besides z of this work, the ones associated with broken bonds in lattice models [21] while those in the second group include the size exponents in Table I of Ref. [3], the roughness exponent [21], and the relaxation exponent z for $I > I_b$ (see below). A disorder-dependent exponent is usually derived from measurements involving a *single* sample, and, hence, fluctuations in its values possibly reflect lack of self-averaging property.

To see how well Eq. (1) represents the relaxation time data, one notes that plotting $(\tau_h/\tau_{ho})^{1/z}$ against I^2/I_b^2 should lead to data collapse. This is indeed seen in Fig. 4 (open symbols, $I^2/I_b^2 < 1$). The solid line is a fit to (1), which is excellent even at low currents. Thus, Eq. (1) holds not only near I_b but also for the entire range of current $I < I_b$. The goodness of the data collapse near I_b is highlighted in the plot in the inset. Straight lines have slopes of unity. To complete the description of the dynamic breakdown, time-to-failure data (closed symbols) obtained above the breakdown point ($\epsilon > 0$) by Lamaignere *et al.* [8] are also shown in the figure. In this case, τ_{ho} is fixed arbitrarily to display the data within the graph and cannot be interpreted as the relaxation time at zero current as in the case $\epsilon < 0$. The dashed line is again a fit to (1) with $z = 1$. The single function fits all the data reasonably well whereas previously, different functions were used to fit different ranges of data. The function $\tau_h \sim (I/I_b - 1)^{-2}$ as considered in Ref. [8] near I_b is also shown in Fig. 4 (dotted line). As seen, it does not fit the whole set of data. It is important to note that the value of the exponent z (i.e., 1) for $\epsilon > 0$ is *independent* of disorder in contrast to the varying exponent for $\epsilon < 0$. This is ensured by Eq. (1) and the requirement that τ_h must vary as I^{-2} at large currents [8] as dissipation of heat becomes negligible at large currents.

It is now useful to summarize characteristics below and above the breakdown point $I = I_b$. (i) For $\epsilon < 0$, the resistance ratio R/R_o (see, for example, Fig. 1 of Ref. [9]) smoothly increases with current to a limiting value (or breakdown ratio) $\Upsilon \approx 1.5$ [16] at $\epsilon = 0$. $R = R(t \to \infty)$ is the steady resistance at a given current. It is best fitted by an expression $R/R_o = \Upsilon + a_1\epsilon + a_2\epsilon^2 + a_3\epsilon^3$ that has a finite slope a_1 at $\epsilon = 0$. For $\epsilon > 0$, there is no steady state. On the other hand, τ_h diverges as a power law of ϵ on both sides. Thus, we have a situation where a susceptibility (e.g., resistance) and a dynamic response variable (e.g., relaxation time) behave *differently* with the driving current in the *same* system and range of disorder. (ii) z is not the same on both sides as discussed above. This is in contrast to equality of the exponents on both sides, which is expected from scaling hypotheses in both

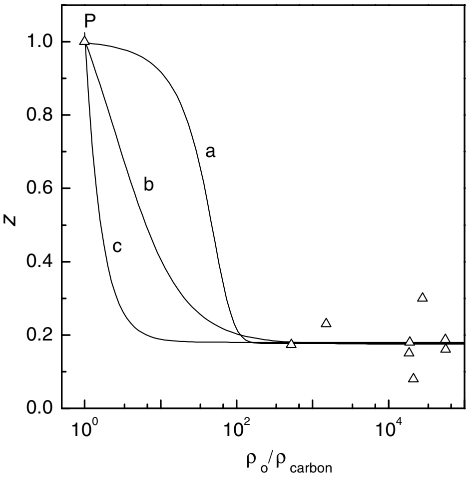

FIG. 3. The exponent z vs the disorder parameter $r = \rho_o/\rho_{\text{carbon}}$ for $I < I_b$. The point P at (1, 1) is a theoretical one corresponding to the pure conductor. The three curves indicate some possibilities for z approaching 1 at P. See text for further discussion.

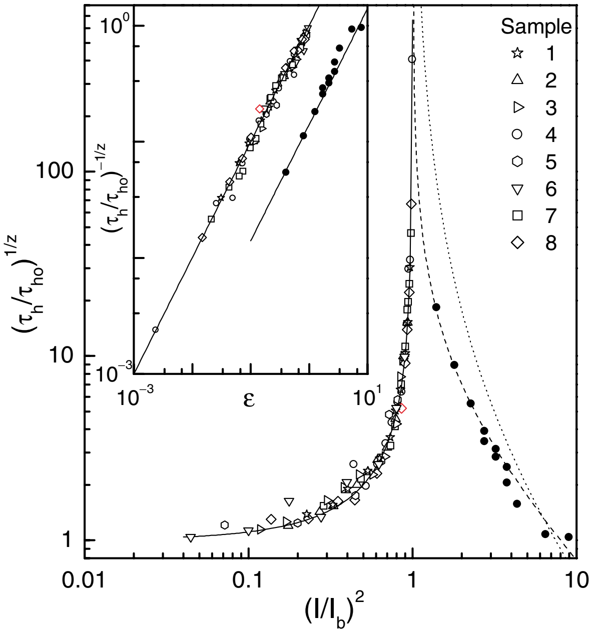

FIG. 4 (color online). Scaling behavior of relaxation time below and above the breakdown point at $I = I_b$. The solid and dashed lines are fits to $(\tau_h/\tau_{ho})^{1/z} = (\mp\epsilon)^{-1}$, respectively. The data corresponding to solid symbols are from Ref. [8].

static and dynamic critical phenomena in thermodynamic systems. (iii) While the scaling in Fig. 4 is itself an expression of "universality" [3], the fluctuations and variation (see below) in z for $\epsilon < 0$ renders the notion of a universality class untenable. (iv) At $\epsilon < 0$, both R and τ_h are *reversible* with respect to current. This rules out any local breakdown *before* the global breakdown occurs. Any irreversible change in the microstructure should also lead to irreversibility in those quantities. This is incompatible with the picture that emerges in breakdown models with *quenched* disorder [2–7,22] where the global breakdown is preceded by increasing bursts of irreversible bond breaking. The composite samples can be considered belonging to a system with quenched disorder for $\epsilon < 0$, and a system with annealed disorder for $\epsilon > 0$. In view of all these, it is difficult to avoid the conclusion that the usual thermodynamic classification of phase transition is inadequate to describe breakdown phenomena.

The presence of disorder is expected to have a profound effect on the breakdown processes. One of its manifestations lies in the conceptual difficulties of taking the limit of disorder going to zero [6]. This is aptly illustrated in the present case. Let us first consider divergence of the relaxation time in a homogeneous medium corresponding to zero disorder. In fact, it has long been discussed [23], albeit couched in the language of stability. The Joule heating under a constant current I in a medium with a positive temperature coefficient of resistance β leads to either a steady state or breakdown depending on whether the heat generated is removed quickly enough or not. Full solutions of heat flow have been obtained in some regular geometries [23] where τ_h is given by

$$\tau_h \sim l^2(\kappa a - bI^2)^{-1} \sim l^2(1 - I^2/I_b^2)^{-1}, \qquad (2)$$

where $I_b^2 = \kappa a/b$, κ is thermal diffusivity, and l is the smallest distance for flow of heat. a, b are constants that depend on the boundary conditions and various material constants including β. Clearly, $z = 1$ in the case of zero disorder ($p = 1$). In passing, let us note that according to Eq. (2), I^2 rather than I is the proper variable to use in problems involving the Joule heating. Consideration of temperature-coupled resistance is an essential ingredient in obtaining (2). Its absence in the dynamic fuse model of Sornette *et al.* [4] gives rise to a relaxation time *independent* of current.

As the disorder is reduced ($p \rightarrow 1$), the relaxation exponent increases from ~0.18 to 1. Thus, the dependence of z on disorder is much stronger than the fluctuations in its values suggest. Presently, there is no theory of z for $p < 1$. An interesting question arises at this point as to how z from a lower value at higher disorder would approach 1 at point P in Fig. 3. Three possible curves are shown schematically. Curve a is intuitive and has a zero slope at P. This means that the system having a small disorder can be simply considered as a homogeneous one with an effective thermal diffusivity. This may be possible if the melting point of the conductor is *less* than that of the insulating matrix. Curve c corresponds to the situation where even a slight disorder leads to an abrupt fall in z. This may happen if the melting point of the conductor is *greater* than that of the insulating matrix. Curve b has a finite slope at P. Clearly, further theoretical efforts are necessary for the understanding of z as a function of disorder. A similar problem [9] concerning the breakdown ratio Υ exists near $p \sim 1$.

In conclusion, a detailed description of the dynamic electric breakdown driven by external current, both below and above the breakdown current, was presented. It was shown that the classification of thermodynamic phase transitions is inadequate for breakdown transitions. The present models having quenched disorder lack reversibility found in composites.

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