Critical Behaviour of Thermal Relaxation in Disordered Systems

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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 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 \(\tau_h\). 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 non-thermodynamic phase transitions.\(^1\) Much of the efforts to understand such irreversible phenomena naturally use 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.\(^2\) 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 similar but somewhat different model, Andersen et al.\(^3\) 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 presence of a tricritical point. On the experimental side, some recent results on electrical failure in composites suggest rather conflicting scenarios. When a sufficiently large current \(I\) is passed through a composite sample (a random binary mixture of a conductor and an insulator\(^4\)) with the conducting fraction \(p\) typically far above the percolation threshold \(p_c \ll p\), Joule heating causes the sample resistance to increase. If the current is more than the breakdown current \(I_b\), heating eventually leads to an irreversible electric breakdown. It was found\(^5\) that the resistance of such a sample increases as a power of time, \(R \sim (\tau_h (I - t)^{-0.65}\) when a constant current \(I > I_b\) is applied. The relaxation time (i.e. time-to-failure) \(\tau_h\) also exhibits a critical behaviour (see below). However, measurements starting with small currents show that the sample conductance drops to zero as soon as the current equals or exceeds the breakdown current, thereby signalling a first-order transition.

In order to resolve this and other issues it would be useful to obtain information about other relevant quantities near the transition. In this paper, we report the results of our dynamic measurements of thermal relaxation using currents less than, but up to, the breakdown cent of the well-known phenomena of slowing down near critical points in thermodynamic systems. The relaxation times also exhibits an unusual scaling relation due to a strong dependence on disorder. The present results together with the earlier ones\(^6\) 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\(^7\) and strain-relaxation measurements in metal networks near the percolation threshold.\(^8\)

Measurements were performed in composites of carbon and wax (C-W) under constant dc current condition at room temperature and the sample resistance was monitored as a function of time during heating. The preparation and characterisation of the system have been described earlier\(^9\). \(p_c\) in C-W system is 0.76\% (by volume). In the tunneling regime\(^10\), 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 \(p_f\) above, which the resistance always increases with bias.\(^11\) The nominal carbon fractions of the samples ranging from 4.5 to 10\% were above or near \(p_f\) which is approximately 4.5\%.\(^12\) Samples prepared with larger carbon fractions were mechanically unstable. On the other hand, samples with lower carbon fractions had tunneling effect offsetting the effect of Joule heating (see Fig. 1 of Ref. 4). The geometry of current flow in a cylindrical 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 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
TABLE I: Various properties of the samples used. $p$ is the carbon fraction by volume (%). $R_o$, $\rho_o$ and $\tau_ho$ are the resistance, resistivity and relaxation time at zero current. $I_b$ is the breakdown current and $z$ is the exponent in Eq. (1).

| Sample No. | $p$ | Height (mm) | $R_o$ (Ω) | $\rho_o$ (Ωcm) | $\tau_{ho}$ (sec) | $I_b$ (mA) | $z$ |
|------------|-----|-------------|-----------|----------------|------------------|-----------|-----|
| 1          | 7.5 | 6.0         | 422       | 567            | 495              | 31.5      | 0.175 |
| 2          | 4.5 | 4.1         | 287       | 561            | 145              | 19.2      | 0.187 |
| 3          | 7.5 | 4.1         | 139       | 290            | 168              | 73        | 0.30  |
| 4          | 4.5 | 2.7         | 77        | 214            | 455              | 83.1      | 0.076 |
| 5          | 7.5 | 2.8         | 73        | 191            | 113              | 112       | 0.18  |
| 6          | 7.5 | 4.2         | 94        | 188            | 156              | 95        | 0.15  |
| 7          | 7.5 | 2.8         | 184       | 15.2           | 67               | 18.7      | 0.23  |
| 8          | 10  | 8.3         | 8.3       | 5.56           | 5.26             | 194       | 135   | 0.173 |

$^a$Nominal value.

$^b$This sample is ribbon-shaped but all other samples are cylinders of 10mm diameter.

current, not exceeding the breakdown current. The relaxation during cooling from a hot steady state was also measured. It exhibited an anomalous behaviour. This will be reported elsewhere.

Figure 1 shows a typical relaxation curve (a) during heating, obtained from a cylindrical sample (#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, $\tau$ is a relaxation time constant and $\alpha$ 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 behaviour. 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 $\tau \propto t^z$. For samples with large $p \gg p_c$ such as the ones used here, the cross-over times should be quite small. Close to $p_c$, $\alpha$ 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. The relaxation time in several samples of different compositions and dimensions (see Table I) are shown in Fig. 2. $\tau_h$ of a sample was found to diverge as a power-law

$$\tau_h = \tau_{ho} |\epsilon|^{^-z} \quad (1)$$

Here, $\tau_{ho}$ is a prefactor, $\epsilon = (I^2/I_b^2 - 1) < 0$ and $z$ is a dynamical exponent. $\tau_{ho}$, $I_b$ and $z$ were treated as fitting parameters and their values are listed in Table I. $\tau_{ho}$ 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, $\tau_{ho}$ diverges as $p$ approaches $p_c$. Such disorder-induced divergences are sometimes attributed to a stretched exponential at short times to a simple exponential at long times $\tau \propto t^z$. For samples with large $p \gg p_c$ such as the ones used here, the cross-over times should be quite small. Close to $p_c$, $\alpha$ 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. The relaxation time in several samples of different compositions and dimensions (see Table I) are shown in Fig. 2. $\tau_h$ of a sample was found to diverge as a power-law

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with $R_o$ as $I_b \sim R_o^{-0.44}$. $R_o$ 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
disorder \((p = 1)\) and increases monotonically with disorder as \(p\) decreases from 1. Within the range of disorder (two orders of magnitude) investigated in this work, the exponent has an average value of \(0.184 \pm 0.06\) i.e. 33% uncertainties. The highest value (0.3) is about four 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: 1) disorder-dependent and 2) disorder-independent or 'universal'. Examples of the first group include, besides \(z\), some of the exponents associated with broken bonds in lattice models \([21]\) while those in the second group include the size exponents in Table I of Ref. \([3]\), roughness exponent \([21]\), 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 show how well Eq. \(\text{[II]}\) represents the relaxation time data, one notes that plotting \((\tau_h/\tau_{h0})^{1/z}\) 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 \(\text{[II]}\), which is excellent even at low currents. Thus, Eq. \(\text{[II]}\) holds not only near \(I_b\) but also for the entire range of current \(I < I_b\). 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. \([\text{5}]\) are also shown in the figure. In this time at zero current as in the case \(\epsilon < 0\). The dashed line is again a fit to \(\text{[II]}\) with \(z = 1\). The single function fits reasonably well all the data 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. \([\text{3}]\) 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. \(\text{[II]}\) and the requirement that \(\tau_h\) must vary as \(I^{-2}\) at large currents \([\text{3}]\) 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 \(R/R_0\) (see, for example, Fig. 1 of Ref. \([\text{1}]\)) smoothly increases with current to a limiting value (or breakdown ratio) \(\Upsilon \approx 1.5\)\([\text{14}]\) at \(\epsilon = 0\). \(R = R(t \to \infty)\) is the steady resistance at a given current. It is best fitted by an expression \(R/R_0 = \Upsilon + a_1\epsilon + a_2\epsilon^2 + a_3\epsilon^3\) that has a finite slope \(a_1\) at \(\epsilon = 0\). For \(\epsilon > 0\), there is no steady state. On the other hand, \(\tau_h\) diverges as a power-law of \(\epsilon\) on both sides; ii) \(z\) is not same on both sides as discussed above. This is in contrast to equality of the exponents on both sides, that is expected from scaling hypotheses in both static and dynamic critical phenomena in thermodynamic systems; iii) While the scaling in Fig. 4 is itself an expression of 'universality' \([\text{2}]\), the fluctuations in \(z\) for \(\epsilon < 0\) renders the notion of an universality class untenable; iv) At \(\epsilon < 0\), both \(R\) and \(\tau_h\) are reversible
versible change in the microstructure should also lead to irreversibility in those quantities. This is incompatible with the picture that emerges in breakdown models with annealed disorder [2, 3, 4, 5, 6, 7] 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 been long discussed [22] 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 \( \beta \) 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 [22] where \( \tau_0 \), is given by

\[
\tau_b \sim I^2(\kappa a - b I^2)^{-1} \sim I^2(1 - I^2/\kappa b)^{-1}
\]  \( (2) \)

where \( I_b^2 = \kappa a/b \), \( \kappa \) is thermal diffusivity, and \( l \) is the smallest distance for flow of heat. \( a, b \) are constants that depends on the boundary conditions and various material constants including \( \beta \). 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 Eq. \( 2 \). 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 exponen increases from \( \sim 0.18 \) 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 the point P in Fig. 3. Three possible curves are shown schematically. The 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. The curve \( c \) corresponds to the situation where even a slight disorder leads to an abrupt fall in \( z \). This may happen if

\[ \epsilon = \frac{\kappa b}{\kappa a} \]

at \( P \). Clearly, further theoretical efforts are necessary for understanding of \( z \) as a function of disorder. Similar problems concerning the breakdown ratio \( \Upsilon \) 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 annealed disorder lack reversibility found in composites.

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