Predictable Electrical Breakdown in Composites

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(Received 25 May 1999)

The Joule regime at large electric fields in composites is presented in the context of a conduction phase diagram in the field-concentration plane. A sample suffers breakdown when the field is too large. The resistance up to breakdown is described by a universal curve as a function of field. It is found that the ratio of the breakdown resistance to the zero-field resistance assumes a fixed value Υ at breakdown. Υ is found to be 1.37 in carbon high-density polyethylene composites and is independent of carbon fraction and external conditions but depends on the nature of the conductor. A quantity which is independent of conducting material is defined. Results are compared with previous data.

PACS numbers: 72.80.Ng, 05.70.Jk, 72.20.Ht

Application of finite force (F, mechanical or electrical) in disordered systems usually results in a nonlinear response leading to some sort of catastrophic behavior in the extreme limit (e.g., fracture in mechanical systems and dielectric breakdown or burning in electrical systems). In recent years there has been a renewed interest in the problem of catastrophic phenomena [1] although the problem of non-Ohmic electrical conductivity in the precatastrophic regime in various disordered systems has been studied for a long time [2,3]. However, there have been very few attempts so far to describe the behavior of a system over the full range of the applied force. Such a study holds the promise of unraveling many important aspects such as precursor effects, predictability, and the effect of disorder on the nature of breakdown. Yagil *et al.* [4] carried out somewhat limited measurements of I-V curves in thin semicontinuous metallic films of Ag and Au. Focusing on breakdown events, it was concluded that breakdown currents I_b in the films scale as $I_b \sim B^{-x}$, where B is the normalized third harmonic component (see below) generated as a result of Joule heating. Breakdown was assumed to occur when the sample resistance R exhibited the first irreversible discontinuity as a function of applied current I. The exponent x was measured to be 0.48 and 0.41 in films of Ag and Au, respectively. The authors also derived theoretical bounds for x, $0.5 \geq x \geq 0.5[1 - 1/t(2 + w_J)]$ so that breakdown currents were expected to lie between two bounds. Here, t is the electrical conductivity exponent and $w_J = \kappa/t$, κ being the noise exponent [5].

In this Letter, we present systematic measurements of electrical resistance, particularly in the Joule regime of a composite system of carbon high-density polyethylene (HDPE) up to breakdown. The breakdown in a sample has the nature of a first-order transition: as soon as the current from a constant current source exceeds a certain value I_b, the sample resistance R starts increasing uncontrollably and becomes unsteady. Let $R_o = R(0)$ be the linear or zero-field resistance, $R_b = R(I_b)$ be the breakdown resistance and $\Upsilon = R_b/R_o$. It is found that for $p > p_J$, where p is the (volume) fraction of conducting component (carbon) and p_J is a fraction characteristic of the system in hand (see below), the ratio of breakdown resistance to linear resistance Υ is a constant which is independent of p, sample geometry and environmental conditions but depends on the nature of the conducting component. This result is quite significant from the point of view of predictability of failures in real materials. Physically, this follows from the observation that the resistance at a given p for all currents up to breakdown follows a simple scaling relation

$$R(I)/R_o = g(I/I_o) \tag{1}$$

and that the breakdown current I_b is proportional to I_o, the current scale for nonlinearity due to Joule heating:

$$I_b \sim I_o\,. \tag{2}$$

These two relations ensure a constant value for Υ at breakdown for $p > p_J$, as observed. Here g is a scaling function. For $I < I_o$, the scaling function $g \approx 1$. For $I > I_o$, $g > 1$. Thus, the current I_o alternatively represents a crossover or onset current which separates the linear regime from the nonlinear regime along the current axis. The same description holds good if field F, instead of current, is used in Eq. (1) with $F_o \sim I_o R_o$.

Previous measurements by Lamaignere *et al.* [6] of the time to failure in a 3D composite system using currents greater than I_b have provided evidence of the critical nature of electrical breakdown. The present mode of breakdown, where the response function R changes suddenly from a finite value to a very large one, is to be contrasted with the phenomenon of the mechanical fracture where elastic moduli go to zero continuously as power laws. It has been suggested that the ratio of two elastic moduli may approach a universal value near fracture [7]. Recently, there have been other suggestions to observe precursor effects [8]. However, to our knowledge, no experiment has been performed thus far to verify these ideas.

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All measurements were done on samples of carbon-HDPE composites corresponding to eight different carbon fractions p. Details of sample preparation and characterization have been given elsewhere [9]. The percolation threshold p_c is 0.17 and $t = 2.9$. The large value of t and resistivity ρ ($\sim 10^{-2}$ Ω cm) of the conducting component (carbon black) in the present system compared to others such as carbon wax ($t = 2$) [3] or Ag film [4] ($\rho \sim 10^{-6}$ Ω cm) makes the Joule effect larger and the regime accessible in a convenient range of p i.e., above $p_J \approx 0.21$. Samples were originally prepared in the form of ribbons with a width of 10 mm and thickness of 1 mm. However, measurements of field-dependent resistances up to breakdown were done at room temperature on samples of sizes $10 \times 5 \times 1$ mm^3 with dc currents from a voltage-limited constant-current source (Keithley 224) flowing parallel to the longer side. A computer was used to acquire data. For simplicity and convenience of comparison, data are discussed below in terms of fields rather than currents.

It is known that, for p close to p_c and for small fields, composites are in the tunneling regime, where $dR/dF \leq 0$, i.e., R initially decreases with field F [3]. This is also seen in Fig. 1 for the sample with nominal concentration $p = 0.2$. However, for large enough field ($F \sim 46$ V/cm), $dR/dF > 0$, i.e., R starts increasing again due to Joule heating until the sample suffers breakdown at about $F \approx 97$ V/cm. All of the other samples with $p > 0.2$ have $dR/dF \geq 0$ for all fields. These samples also suffer breakdown at sufficiently high field. Breakdown occurs when one of the constituents of a composite sample (insulating HDPE in the present case or conducting metals in thin films [4]) melts locally due to the loss of balance between generation and dissipation of heat as the sample current is increased. Consequently, the sample resistance becomes unstable. Each point in Fig. 1 corresponds to a steady state. Experimentally, the breakdown resistance R_b of a sample is taken to be the steady state resistance measured just before the breakdown and corresponds to the last stable point in its R-F curve. The error in the measured value of R_b is proportional to the last increment in current (ΔI) which leads to an eventual breakdown. The corresponding increment in field $\Delta F = R_b \Delta I$ applied to each sample is indicated in Fig. 1. Increments in fields, although small, have an amplifying effect on materials with a positive temperature coefficient of resistance under constant current supply. It is seen that Υ for all samples varies within a small range. Considering breakdowns with smaller ΔF, we take $\Upsilon \approx 1.37$. This ratio also agrees with values obtained from limited measurements done by passing currents under constant voltage. Note that some (◇, △) of the samples are used for the first time while others have previously suffered breakdown more than once. Also, Fig. 1 contains data from two samples with the same $p = 0.4$ but of different lengths (×, 1 cm; ◇, 1.8 cm). This shows that the ratio Υ is independent of geometry or initial conditions. The robustness of Υ is further demonstrated in the inset of Fig. 1 which displays similar data but for only one sample ($p = 0.4$, ◇ in Fig. 1) with different cycle numbers (n) as indicated. The sample resistance always increases after a breakdown cycle. These results are summarized in the conduction phase diagram in the F-p plane (Fig. 2). The curve a separates the linear regime ($dR/dF \simeq 0$) from the tunneling regime and meets the curve b at a point "J," to be called the "Joule point" corresponding to $p = p_J \approx 0.21$ and $F = F_J \approx 5.7$ V/cm. The curve b separates the linear regime from the Joule regime for $p > p_J$ and the tunneling regime from the Joule regime for $p < p_J$. There is no tunneling regime ($dR/dF < 0$) for $p > p_J$. Note that measured points both below and above p_J are fitted by the same function. The point J represents a multicritical point in analogy with the thermal phase transitions. All of the three curves represent transitions of a continuous nature, in contrast to the breakdown curve c which is first order in nature. The portion of the curve c for $p > p_J$ is characterized by a constant $\Upsilon \approx 1.37$ [10].

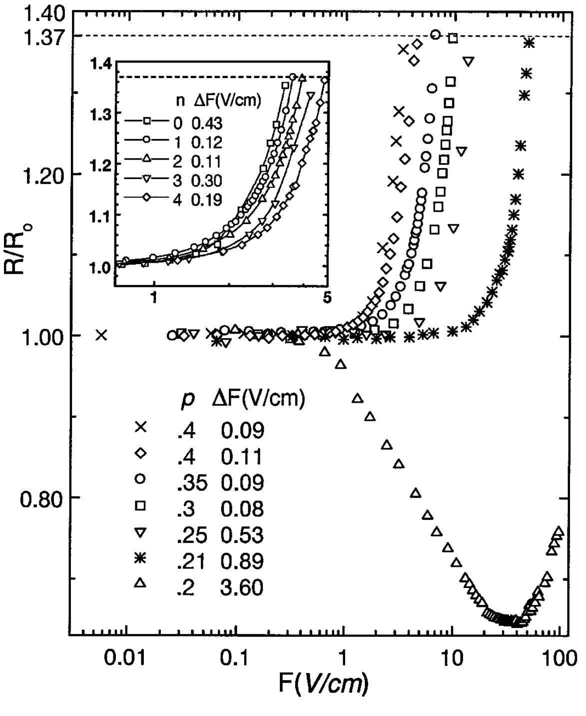

FIG. 1. Semi-log plot of the ratio of resistance R and its zero-field value R_o as a function of the field for different samples of carbon-HDPE. The carbon volume fraction p of each sample is as indicated. See text for explanation of ΔF. Inset: plots of R/R_o vs field F for the sample with $p = 0.4$ but different breakdown cycles. The cycle numbers (n) are as indicated. The solid lines are only guides to eyes.

Figure 3 shows a plot of normalized resistance R/R_o for each curve in the Joule regime in Fig. 1 against the scaled field F/F_o. It is seen that all curves collapse onto a single curve right *up to* the breakdown. This verifies

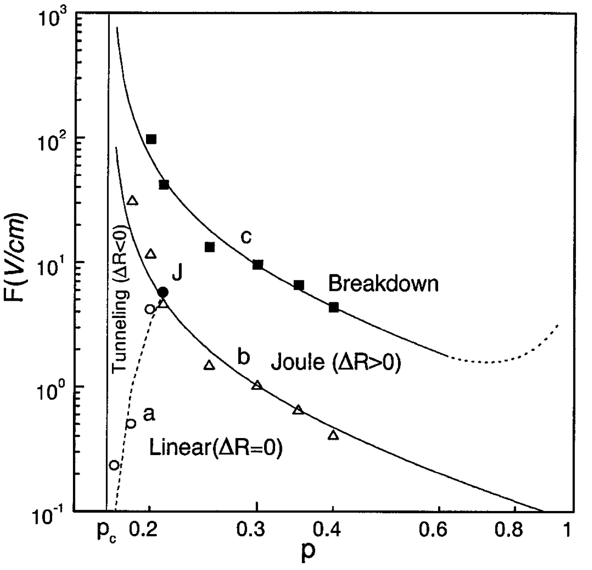

FIG. 2. Conduction phase diagram in the field-fraction (F-p) plane of the carbon-HDPE composite. ΔR is change of resistance due to an increase in field. See text for explanation of the point "J". Solid lines are fits according to $F = 0.065R_o^{.45}$ (lower) and $F = 0.6R_o^{.44}$ (upper) where R_o is the zero-field resistance of a sample. The dashed lines is a guide to eye.

Eq. (1). A log-log plot of onset field F_o vs R_o is shown in the inset of Fig. 3. The straight line fit indicates that F_o scales with R_o as $F_o \sim R_o^{y_o}$ with the onset exponent $y_o = 0.45 \pm 0.01$. A plot of the breakdown field F_b vs R_o is also shown in the same inset. Clearly, F_b also scales with R_o as $F_b \sim R_o^{y_b}$ with the breakdown exponent $y_b = 0.44 \pm 0.01$. Thus, $y_b \approx y_o$, which supports Eq. (2). This result can be easily understood if considered in the spirit of mean-field theory. Let ΔT be the average temperature rise caused by Joule heating due to current I. The change in resistance ΔR is then given by $\Delta R = \beta_M R_o \Delta T = \beta_M R_o h_M I^2 R_o$. Here $\beta = (1/R)\Delta R/\Delta T$ is the macroscopic temperature coefficient of resistance, and h_M is the ratio between temperature rise ΔT and power generated in the sample, $I^2 R_o$ [11]. The onset current scale is given by the condition that $\Delta R \sim R_o$. This gives $I_o \sim R_o^{-1/2}$ so that, with $F_o \sim I_o R_o$, $y_o = 0.5$. The breakdown may be defined by the condition that ΔT must reach some higher value ΔT_M corresponding to the melting of one of the components (HDPE in this case). This leads to $\Delta T_M = h_M I_b^2 R_o$ so that $y_b = 0.5$. Hence, $I_b \sim I_o$. For a random resistor network, the change in resistance due to Joule heating, in the first approximation, $\Delta R = R - R_o$ is given by $a\beta h R_o^2 S I^2$ [12]. Hence,

$$R = R_o + a\beta h R_o^2 S I^2. \tag{3}$$

Here a is a simple constant. β and h are the same as before but now need to be defined in terms of a single resistance element. S is the relative noise power being proportional to the fourth moment of the current distribution and is defined by $S \sim R_o^{w_J}$ with w_J defined earlier. The noise for the samples for $p > p_J$ has been measured (details will be published elsewhere) and are shown in the inset of Fig. 3 (closed symbol). It is seen that $w_J \approx 0.13$ which is much less than that found near p_c [5,13] (hence the deliberate use of the suffix J in w_J to emphasize the fact that the regime of concern corresponds to $p \gg p_c$). Upon comparison of (3) with (1), it is seen that the scaling function $g(z)$, to a first approximation, is given by $g(z) \simeq 1 + z^2$ and $I_o \sim (R_o S)^{-1/2}$. The latter, along with $F_o \sim I_o R_o$, yields $y_o = (1 - w_J)/2 \approx 0.44$ which agrees well with the experimental value. To find scaling for I_b, we note that the singly connected bonds (SCBs) are the ones that will see a maximum rise in temperature causing adjacent HDPE to melt first [4]. Let I_{SCB} be the average current through each SCB so that the average temperature rise is $\Delta T = h r_o I_{\mathrm{SCB}}^2$ where r_o is the typical resistance of a single bond. Now, $I_{\mathrm{SCB}} \sim \xi^{d-1} I \sim R_o^{(d-1)\nu/t}$ where ξ and ν are the correlation length and exponent, respectively, and d is the dimensionality. As the current is increased, ΔT will rise eventually to some ΔT_m when a SCB melts. Since the actual breakdown current I_b is most likely to be less than the average current, we have $I_b \leq R_o^{-(d-1)\nu/t}$ so that, with $F_b \sim I_b R_b \sim I_b R_o$,

$$y_b \leq 1 - (d - 1)\nu/t. \tag{4}$$

With $\nu = 0.9$ and $t = 2.9$ for 3D, $y_b \leq 0.4$ which is consistent with the experimental value of 0.44 within errors.

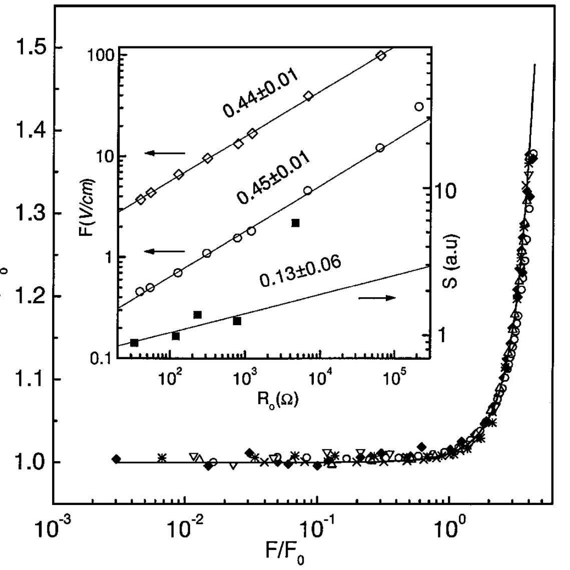

FIG. 3. Scaled plot of normalized resistance vs scaled field of the data in the Joule regime of the Fig. 1. The solid line is a fit to the data according to $y = 1 + 0.01x^2 + 0.0009x^4$. Inset: Log-log plots of F_o-R_o (°), F_b-R_o (◇) and S-R_o(solid square). The solid lines are the power law fits to the data with the exponents as indicated.

Let us consider the theoretical values of the exponents. Far above the percolation threshold ($p \gg p_c$), the effective medium theory [14] predicts $w_J \approx 1$. Also, $(d-1)\nu/t$ is 1 and 0.9 in 2D and 3D, respectively. Thus, both y_o and the upper bound of y_b are close to zero, making Eq. (2) plausible. It is remarkable that in the present case (3D) Eq. (2) is satisfied despite large deviations of observed values of w_J and t from their theoretical ones. In 2D films, Yagil *et al.* [4] measured B rather than I_o. Since B is defined by $R = R_o + BI^2$ it follows from (3) that $B \sim R_o/I_o^2$. This gives $y_o \approx -0.1\ (-0.3)$ and $y_b \approx -0.7\ (-0.7)$ for Ag (Au) film. It appears that the relation (2) may hold good within errors at least for Au but not for Ag. On the other hand, x is calculated to be 0.24 in carbon HDPE. This value is outside the bounds for x in 3D that can be derived by the following [4]: $0.5 \geq x \geq 0.5\{1 - [1 + (d-2)\nu]/t(2 + w_J)\}$. After putting values we obtain $0.5 \geq x \geq 0.36\ (0.35)$, where $t = 2\ (2.9)$ and $w_j = 1.5\ (0.13)$. The values in brackets are as observed. Interestingly, if the *measured B* is taken to be proportional to I_o^{-2}, instead of R_o/I_o^2, then $I_o \sim B^{-0.5} \sim R_o^{-1.6(1.8)} \sim I_b$ for Ag (Au) film. In this case, $I_b \sim I_o \sim B^{-0.5}$ even in carbon HDPE satisfying the bounds ($2t$ is to be replaced by t in the lower bound).

$\Upsilon = R_b/R_o$ is dependent on the nature of the conducting component. From (3) it is seen that $\Upsilon - 1$ would be proportional to $\beta h \rho_o$, where ρ_o is the resistivity of the conductor. h, which controls heat transfer, is proportional to τ, the thermal conductivity. These lead us to expect that the quantity Λ defined by

$$\Lambda = (\Upsilon - 1)/\beta\tau\rho_o$$

would have a universal value at breakdown for $p > p_J$. This is borne out by the data of carbon HDPE and Ag films [4]: for carbon HDPE, $\Upsilon = 1.37$, $\beta \sim 10^{-3}\ \mathrm{K}^{-1}$ [15], $\tau = 0.016\ \mathrm{W\,cm^{-1}\,K^{-1}}$ [16] and $\rho_o \sim 10^{-2}\ \Omega\,\mathrm{cm}$ [9]; for Ag the corresponding values are 1.016, 0.004, 4.29, and 1.6×10^{-6}. In both cases, $\Lambda \approx 10^6\ \mathrm{W^{-1}\,cm\,K^2}$. It is now possible to explain the breakdown in the tunneling sample in Fig. 1 ($p = 0.2$). Let us take Υ as the ratio of the breakdown resistance and the minimum resistance (~ 1.2) and the effective conductor resistivity in the minimum resistance state as 0.63 times the carbon resistivity, $10^{-2}\ \Omega\,\mathrm{cm}$. Hence, $(\Upsilon - 1) \times 10^{-2}/\rho_o \approx 0.2/0.63 = 0.32$ which is close to 0.37 as in the case of other samples without any tunneling regime. Incidentally, the break in slope of I_b vs R_o in Au films [4] may be due to the inclusion of data both above and below p_J.

Finally, a comment about the role of disorder [17] in the process of breakdown is in order. Consider the case where the conductor has a much higher melting temperature than the insulator as in the present case. Near $p = 1$, one would expect the curve c in Fig. 2 to rise upwards, as indicated by the dotted line, without any change in the nature of breakdown. In the case where the conductor melts at a lower temperature, the curve c continues towards $p = 1$ as per the fit but with y_b tending to $1/2$. It is interesting to note that the scaling relation such as (1) has been observed in many disordered systems including ones with hopping conduction [18]. However, a Joule system has the virtue of its mechanism being understood, whereas the mechanism of nonlinear conduction in hopping systems is far from being clear.

We acknowledge discussions with B. K. Chakrabarti and assistance of Arindam Chakrabarty in data acquisition and processing.

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[10] Note that the "tunneling" regime in Fig. 1 refers only to the macroscopic behavior of resistance. However, microscopically tunneling may take place even in the "linear" regime. See also Ref. [13].

[11] The quantity h_M incorporates the effects of external conditions such as cooling rate etc. In an experiment where a fan was used to cool the sample the value of I_o which depends on h_M increased from the value corresponding to the case where no fan was used, but Eqs. (1) and (2) remained intact.

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