# Universal scaling and nonlinearity in surface layer fragmentation

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We analyze disordered one-dimensional bond-network models and investigate the influence of nonlinear force-elongation relations $F \propto |x|^m$ on the fragmentation of coatings under tension. We aim to elucidate the interplay between the nonlinear forces and the random failure thresholds of the bonds. After an initial stage, the mean fragment length $\langle L \rangle$ scales with the applied strain $\varepsilon$, $\langle L \rangle \propto \varepsilon^{-\alpha}$, where $\alpha = m/(m+1)$ for weakly disordered and $\alpha = m/(m+2)$ for strongly disordered coatings.

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Breakage phenomena range from the damage of macroscopic objects such as the destruction of glass windows to the failure on smaller length scales, e.g., the formation of crazes in poly(methylmethacrylate) (PMMA). Because of the technological and scientific interest in fractures, the subject has a long history; recently breaking of coatings has attracted new attention [1–16], particularly the fragmentation of thin brittle films under uniaxial tension. Here cracks once nucleated grow perpendicular to the stress direction and form separate, nearly rectangular fragments, see, e.g., Ref. [12]. This process can be described by a one-dimensional model [17–20], which allows to a great extent an analytical treatment. We focus on the mean fragment length $\langle L \rangle$ as a function of the strain $\varepsilon$ to which the substrate is subjected.

Recent studies [17–20] have investigated the dependence of $\langle L \rangle$ on $\varepsilon$ based on *linear* force-elongation relations, and have revealed a scaling law:

$$\langle L \rangle \propto \varepsilon^{-\alpha}. \tag{1}$$

However, for many materials (such as polymers) linear behavior occurs only in a very restricted range of deformations. Thus in this paper we analyze the influence of *nonlinear* force-elongation relations on fragmentation. Surprisingly, it turns out that $\langle L \rangle$ also scales when the forces are nonlinear.

To fix the ideas, we start from a one-dimensional model [17–22], see Fig. 1. The coating consists of $N$ nodes connected by $N-1$ breakable springs of equilibrium length $l_{\mathrm{eq}}$. Under stress the bonds elongate, which leads to the restoring force $f_k = d u_k$ due to the $k$th surface layer spring; $d$ being the elastic constant and $u_k$ the spring's elongation. The interaction between coating and substrate is modeled by springs of elongation $v_k$ and (non-necessarily Hookean) restoring forces $F_k(v_k)$. These springs are anchored to the substrate at equidistant nodes. For better illustration, in Fig. 1 the bonds between coating and substrate are shifted vertically. If the distance between the substrate's nodes increases from $l_{\mathrm{eq}}$ to $l_{\mathrm{eq}} + \Delta l$ (being a relative elongation of $\varepsilon = \Delta l / l_{\mathrm{eq}}$), the bonds in the coating expand. Increasing $\varepsilon$ continuously corresponds to stretching the substrate uniaxially. In order to mimic the occurrence of cracks in the coating, we assume that the $k$th surface layer bond breaks irreversibly, when its elongation $u_k$ exceeds a random failure threshold $u_b(k)$. The thresholds $u_b(k)$ are chosen randomly at the beginning of the fragmentation process and then are fixed (quenched disorder); they obey a probability distribution, here denoted by $p(u_b)$.

From the arrangement depicted in Fig. 1 a differential equation for the elongations $v(k) \equiv v_k$ follows. In each ''loop''

$$\Delta l = \varepsilon l_{\mathrm{eq}} = v_k - v_{k+1} + u_k \tag{2}$$

holds. Subtracting from Eq. (2) the corresponding equation for $k-1$ leads to

$$v_{k+1} - 2v_k + v_{k-1} = u_k - u_{k-1} = \frac{1}{d}(f_k - f_{k-1}), \tag{3}$$

where we used $f_k = d u_k$. The equilibrium condition for the $k$th layer node is given (in our scalar picture) by

$$F_k = f_k - f_{k-1}. \tag{4}$$

Going to a continuous description, one can replace the left-hand side of Eq. (3) by $v''(k) \equiv d^2 v / dk^2$, so that we are led to

$$v''(k) = \frac{F(k)}{d}. \tag{5}$$

At the boundaries the forces $f_k$ vanish, i.e., $f_0 = f_N = 0$; we may also set $u_0 = u_N = 0$. From this and Eq. (2) one has as boundary conditions $v'(1/2) = v'(N+1/2) = -\varepsilon l_{\mathrm{eq}}$.

Previous studies on the fragmentation of coatings [17,18,20] and of fibers [23,24] have focused on purely lin-

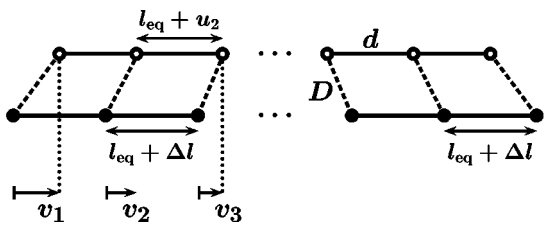


FIG. 1. Bond-network model for fragmentation of coatings under uniaxial tension. Here $d$ and $D$ denote the elastic constants of the two types of springs and $l_{\mathrm{eq}}$ is the equilibrium length. The elongations of the bonds are denoted by $u_k$ ($d$ bond), respectively, $v_k$ ($D$ bond). The bonds between coating and substrate are shifted vertically for better illustration.

 

ear force-elongation relations, respectively, on random fuse networks with linear current-voltage relations [2]. In this context we have previously found that linear one-dimensional (1D) systems reproduce the scaling of linear 2D objects quite satisfactorily [19]. Here, in this paper, we introduce quite general, nonlinear forms for the forces $F(v) \equiv F_k[v(k)]$; we assume that the interaction between coating and substrate is given by a force-elongation relation of the form

$$F(v) = D\,\mathrm{sgn}(v)\left(|v| + \frac{|v|^m}{b}\right), \tag{6}$$

where $D$, $b$, and $m$ are positive parameters ($m>1$). Equation (6) may be viewed as ''stress-hardening,'' as occurs, e.g., in networks containing nonextensible elements, finite extensible nonlinear elasticity (FENE).

To obtain the elongations $u(k) \equiv u_k$, we consider that from Eqs. (3) and (4) we have in the continuum

$$F(k) = d[u(k) - u(k-1)] \approx d[u'(k-1/2)] \tag{7}$$

for the $k$th layer node, which yields

$$u(k) = \frac{1}{d}\int_0^k F(n+1/2)\,dn. \tag{8}$$

We can now readily evaluate $u(k)$ for not too early stages of fragmentation. Because of symmetry, it suffices to consider only the left half of the coating, where $v(k) \geqslant 0$ holds. Hence, from Eq. (5) also $v''(k) \geqslant 0$ for $k \in [1/2,(N+1)/2]$ follows. Therefore the maximal slope of $v(k)$ (largest absolute value) is given at 1/2 with $v'(1/2) = -\varepsilon l_{\mathrm{eq}}$ (boundary condition) and its minimal slope is attained at $(N+1)/2$ with $v'[(N+1)/2] \approx u_{\mathrm{prob}} - \varepsilon l_{\mathrm{eq}}$, as follows from Eq. (2) with $u(N/2) \approx u_{\mathrm{prob}}$, where $u_{\mathrm{prob}}$ is an estimate for $\langle u(N/2)\rangle$ at breakage. Using these results, we can estimate $v(k)$ in the interval $[1/2,(N+1)/2]$ by considering the lower and the upper bound for $v(k)$:

$$\begin{aligned} -(\varepsilon l_{\mathrm{eq}} - u_{\mathrm{prob}})[k-(N+1)/2] \\ \leqslant v(k) \leqslant -\varepsilon l_{\mathrm{eq}}[k-(N+1)/2]. \end{aligned} \tag{9}$$

Equation (9) is the linear approximation of $v(k)$ near $(N+1)/2$, where $v'[(N+1)/2]$ is estimated by $-\varepsilon l_{\mathrm{eq}}$, respectively $u_{\mathrm{prob}} - \varepsilon l_{\mathrm{eq}}$.

The relative deviation of these two limits is $\Delta v/v = u_{\mathrm{prob}}/(\varepsilon l_{\mathrm{eq}})$, so that for $\varepsilon$ large $v(k)$ is nearly $v(k) = -\varepsilon l_{\mathrm{eq}}[k-(N+1)/2]$. Furthermore the $v(k)$ get to be large in the later stages of fragmentation, so that the $|v|^m$ term dominates in Eq. (6). Therefore we find from Eq. (8)

$$u(k) \approx N^{m+1}\varepsilon^m A g_m(z), \tag{10}$$

where we set $z = k/N - 1/2$ and $g_m(z) = 1 - |2z|^{m+1}$. The constant $A$ is given by $l_{\mathrm{eq}}^m/[\xi^2 b(m+1)2^{m+1}]$ and the correlation length $\xi$ is defined by $\xi = \sqrt{d/D}$. From Eq. (10) it follows that an increase in the nonlinearity (i.e., in $m$) changes $u(k)$ from a parabolic shape (for $m=1$) to a plateau shape (for $m$ very large). These considerations are valid both for the initial system and, after replacing $N$ by $L$, for subsequent fragments of length $L$.

Equation (10) leads to a rough estimate for the occurrence of cracks for large $\varepsilon$. If a bond breaks, we have $u(N/2) \approx u_{\mathrm{prob}}$, so that

$$\varepsilon = \left(\frac{u_{\mathrm{prob}}}{A}\right)^{1/m} N^{-(m+1)/m}. \tag{11}$$

holds.

Now let us turn to the fragmentation process. The bonds' elongation $u(k,L) \equiv u_k$ within a fragment of length $L$ attains its maximum in the middle of each segment. The maximum of $u(k)$ increases with larger $\varepsilon$ and larger fragment length $L$. For a fixed value of $\varepsilon$ larger fragments break more readily than shorter ones. We thus introduce $L_c(\varepsilon)$ as the length of a fragment, whose probability to stay intact under strain $\varepsilon$ is 1/2. Now we assume that $L_c(\varepsilon)$ is the only relevant length scale and hence that $\langle L(\varepsilon)\rangle$ is proportional to $L_c(\varepsilon)$. In the following, we show how $L_c(\varepsilon)$ is obtained. We denote by $p(u_b)$ the distribution of the local failure thresholds (ranging from $u_{\min}$ to $u_{\max}$) and by $F_{cu}(u_b) = \int_0^{u_b} p(u)\,du$ the corresponding cumulative distribution function. The probability $P$ for a fragment of length $L$ to stay intact, until the substrate's strain reaches $\varepsilon$, is

$$\begin{aligned} P &= [1 - F_{cu}(u(1))] \times [1 - F_{cu}(u(2))] \\ &\quad \times \cdots \times [1 - F_{cu}(u(L-1))] \\ &= \exp\left\{\sum_{k=1}^{L-1} \ln[1 - F_{cu}(u(k))]\right\}. \end{aligned} \tag{12}$$

As discussed in Ref. [22], only the behavior of $F_{cu}(u_b)$ for $u_b$ close to $u_{\min}$ is essential, most of the $F_{cu}[u(k)]$ are small compared to one, and one has for $P$ by reverting to integration and expanding the logarithm:

$$P \approx \exp\left\{-\int_0^L F_{cu}[u(k,L)]\,dk\right\}. \tag{13}$$

Inserting $P = 1/2$, we are led to

$$\int_0^{L_c} F_{cu}[u(k,L_c)]\,dk = \ln 2. \tag{14}$$

Moreover the change of variable $z = k/L_c - 1/2$ yields

$$L_c \int_{-1/2}^{1/2} F_{cu}[L_c^{m+1}\varepsilon^m A g_m(z)]\,dz = \ln 2, \tag{15}$$

where $g_m(z)$ is a function of $z$ only. From Eq. (15) the scaling laws for different forms of the probability distribution $p(u_b)$ follow. Knowing that the behavior of $p(u_b)$ close to $u_{\min}$ determines the fragmentation kinetics, we assume the following power-law forms for $p(u_b)$:

$$p(u_b) = \begin{cases} \kappa(u_b - u_{\min})^\beta & \text{for } u_{\min} \leqslant u_b \leqslant u_{\min} + W \\ 0 & \text{otherwise} \end{cases} \tag{16}$$

with $\kappa = (\beta+1)W^{-(\beta+1)}$ and $\beta \geqslant 0$. Around $u_{\min}$ Eq. (16) is representative for many other forms in use, e.g., the Weibull distribution. In the case of weak disorder, i.e., for small $W/u_{\min}$ or $\beta$ large, $p(u_b)$ is centered in a small domain of

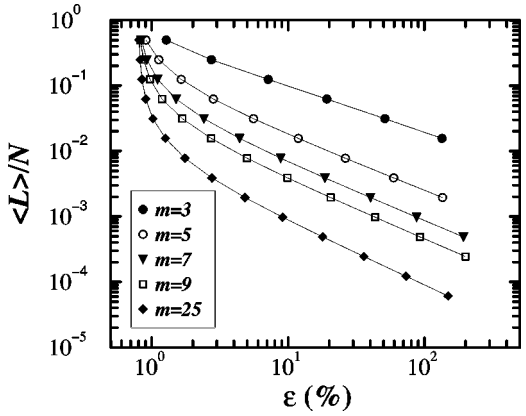


FIG. 2. The function $\langle L(\varepsilon)\rangle$ in homogeneous systems for different values of $m$. The parameters are $N=2^{15}$, $\xi^2/N^2=0.1$, $b=6\times10^4$, $u_b=0.008$, and $l_{\rm eq}=1$. The $m$ values are indicated in the figure.

$u_b$; then the integral in Eqs. (14) and (15) is nonzero only if $L_c^{m+1}\varepsilon^m A g_m(0)\approx u_{\min}$. Thus we find

$$L_c^{m+1}\propto\varepsilon^{-m}, \tag{17}$$

i.e.,

$$\langle L\rangle\propto L_c\propto\varepsilon^{-m/(m+1)}. \tag{18}$$

The case $m=1$, corresponding to a linear spring, reproduces the previously established result $\langle L\rangle\propto\epsilon^{-1/2}$ for the hierarchical fragmentation process [17,18]. Interestingly, not only in the case of purely linear bonds, but also for nonlinear bonds exact scaling solutions exist. In the case of strong disorder, namely, large $W/u_{\min}$ (e.g., $u_{\min}=0$), and $\beta=0$ leading to $p(u_b)=1/W$ for $0\leqslant u_b\leqslant W$, we get $F_{cu}(u_b)=u_b/W$, and $L_c$ is given by

$$L_c^{m+2}\varepsilon^m\frac{AJ}{W}=\text{const}, \tag{19}$$

with $J=\int_{-1/2}^{1/2}g_m(z)dz$, from which it immediately follows that

$$\langle L\rangle\propto L_c\propto\varepsilon^{-m/(m+2)}. \tag{20}$$

Equation (20) reproduces the strong disorder result $\langle L\rangle\propto\varepsilon^{-1/3}$ for linear springs in the case of $\beta=0$ [17,18]. The probability distributions with $u_{\min}=0$ starting in a power-law fashion for small values of $u_b$, i.e., $p(u_b)\propto u_b^\beta$, lead to $F_{cu}(u_b)\propto u_b^{\beta+1}$, and thus

$$L_c(L_c^{m+1}\varepsilon^m)^{\beta+1}=\text{const}, \tag{21}$$

from which $\alpha$ of Eq. (1) follows readily:

$$\alpha=\frac{m(\beta+1)}{(m+1)(\beta+1)+1}. \tag{22}$$

The two previous results, Eqs. (18) and (20), are obtained in this general case as the limits of $\beta=0$ (flat distribution) or $\beta\to\infty$ (concentrated distribution). From Eq. (22) we see that the power-law exponent simply is a function of the nonlinearity characterized by $m$ and the disorder. No other parameters, such as the elastic constants of the materials, influence $\alpha$. Because of $m>1$ the power-law exponent ranges between 1/3 and 1, depending on the values of $m$ and $\beta$. For linear bonds ($m=1$) the maximum value for $\alpha$ is 1/2.

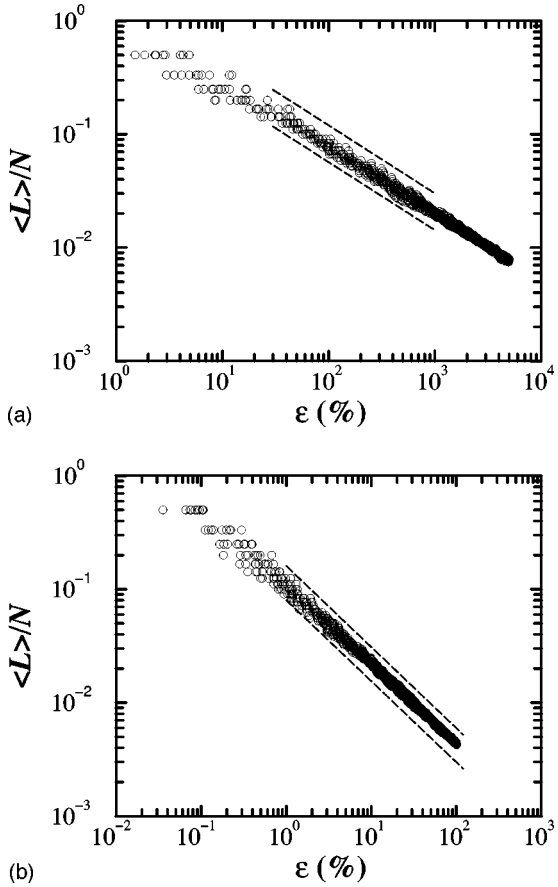


FIG. 3. Mean fragment length $\langle L(\varepsilon)\rangle$ in a strongly disordered system ($u_{\min}=0,\beta=0$) for two sets of parameters. Each curve is obtained by simulations with ten different realizations of the probability distribution Eq. (16). The slopes of the dashed lines are (a) 3/5 and (b) 5/7. Parameters: (a) $N=2^{15}$, $\xi^2/N^2=10^5$, $b=120$, $W=1$, $m=3$, and $l_{\rm eq}=1$. (b) $N=2^{15}$, $\xi^2/N^2=10^3$, $b=120$, $W=0.5$, $m=5$, and $l_{\rm eq}=1$.

The onset of the scaling regime is given by a crossover length $\zeta$ for $\langle L\rangle$. The mean fragment length $\langle L(\varepsilon)\rangle$ scales, when the elongations $v(k)$ are a linear function of $k$. For small segments, the deviation of the linear approximation Eq. (9) and the solution $v(k)$ at 1/2 is roughly given by $v''(1/2)N^2/8$. If this deviation is small with respect to $v(1/2)\approx N\varepsilon l_{\rm eq}/2$, then the crossover takes place. Defining the crossover by, say $Nv''(1/2)/(4\varepsilon l_{\rm eq})=1/8$, we find from Eq. (5), neglecting on the right-hand side the linear term in $v$,

$$\frac{Nv''(1/2)}{4\varepsilon l_{\rm eq}}=\frac{N^{m+1}(\varepsilon l_{\rm eq})^{m-1}}{\xi^2 b 2^{m+2}}=\frac{1}{8}. \tag{23}$$

Using Eq. (11) we obtain for the crossover length $\zeta$

$$\zeta=\left(\frac{\xi^2 b}{[2(m+1)u_{\rm prob}]^{m-1}}\right)^{1/(m+1)}. \tag{24}$$

Hence the scaling regime sets in as soon as $\langle L \rangle$ gets smaller than $\zeta$.

In order to exemplify these findings, we have simulated the fragmentation process for various sets of parameters in systems with $N=2^{15}=32\,768$ bonds. We solve numerically Eq. (5) with the given boundary conditions. Starting with $\varepsilon=0$, we increase incrementally $\varepsilon$. At each step the elongations of the surface layer bonds are given by Eq. (2) in the continuous limit, which is $u(k)=\varepsilon l_{\mathrm{eq}}+v'(k+1/2)$. If $u(k)$ exceeds its specific failure threshold $u_b(k)$, the $k$th layer bond is irreversibly removed. Then for each new fragment Eq. (5) is solved again with the incrementally increased values for $\varepsilon$.

The results of the numerical simulations are shown in Figs. 2 and 3. Figure 2 shows the mean fragment length $\langle L \rangle$ in a homogeneous system [$W=0$ in Eq. (16)] for the parameters $u_b=0.008$, $\xi^2/N^2=0.1$, $N=2^{15}$, and $b=6\times10^4$. The nonlinear force-elongation-relation $F(v)$ is given by Eq. (6) with $m=3$, 5, 7, 9, respectively 25. Least-squares fits of the curves in the final stage ($\varepsilon$ large) show that $\langle L(\varepsilon) \rangle$ decays indeed algebraically with the values of $\alpha$ given by Eq. (18). Moreover we have also performed simulations for two sets of parameters in the strongly disordered case, namely, Eq. (16) with $u_{\min}=0$ and $\beta=0$. The results are plotted in Fig. 3 for the parameters (a) $W=1$, $N=2^{15}$, $\xi^2/N^2=10^5$, $b=120$, $m=3$ and for the parameters (b) $W=0.5$, $N=2^{15}$, $\xi^2/N^2=10^3$, $b=120$, $m=5$. For each set of parameters we have chosen ten different realizations of the probability distribution, so that each of the curves in Fig. 3 corresponds to ten realizations. Once again the mean fragment length scales for large values of the strain. From a least-squares fit we obtain for (a) $\alpha=0.594$ and for (b) $\alpha=0.712$; the corresponding slopes are depicted in Fig. 3 as dashed lines, and the values may be compared to the analytical results $3/5=0.600$ and $5/7\approx0.714$. In conclusion, the simulations agree quantitatively with our analytical considerations.

In summary, we have studied the influence of nonlinear forces on the fragmentation of coatings under uniaxial tension. Our analysis shows that the mean fragment length $\langle L \rangle$ versus applied strain $\varepsilon$ decays as a power law, $\langle L \rangle \propto \varepsilon^{-m(\beta+1)/[(m+1)(\beta+1)+1]}$, where $m$ is the highest nonvanishing power in the force-elongation relation and $\beta$ characterizes the lower end of the failure threshold distribution. For large values of the strain, this power law is exact and only depends on two parameters. Hence the (mesoscopic) determination of $\langle L \rangle$ is a powerful tool in order to determine the microscopic behavior of forces acting in surface layer fragmentation.

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