Disorder and plasticity in the fragmentation of coatings

U. A. Handge,* I. M. Sokolov, and A. Blumen
Theoretical Polymer Physics, University of Freiburg, Hermann-Herder-Strasse 3, 79104 Freiburg im Breisgau, Germany
(Received 28 November 2000; published 14 June 2001)

Using a one-dimensional model that takes into account ideal plasticity of the surface layer, we investigate the fragmentation of thin coatings under uniaxial tension. The coating is modeled as a chain of plastically deforming elements that are connected via leaf springs to a uniformly stretched substrate. Each coating element can only withstand a maximum elongation, which is randomly distributed. From simulations of the fragmentation process we find that the average crack spacing $\langle L\rangle$ scales with applied strain ε, i.e., $\langle L\rangle \propto \varepsilon^{-\kappa}$. Simulations and analytical arguments show that the scaling exponent κ depends on the disorder parameters of the model.

DOI: 10.1103/PhysRevE.64.016109 PACS number(s): 05.40.−a, 46.65.+g, 46.50.+a

Multiple cracking phenomena are ideally suited to study the interplay between the statistical aspects of failure phenomena and the elastic properties of solids [1–3]. Common examples for sequential cracking processes are the fragmentation of thin brittle coatings, the fragmentation of fibers in matrices and matrix cracking in cross-ply laminates [4]. During such fragmentation processes a large number of breakage events occurs, which allows us to use the information obtained from the fragmentation kinetics to analyze the micromechanical properties of the system [5].

Several research groups have investigated the fragmentation of thin brittle coatings on ductile substrates under uniaxial tension, see, e.g., Refs. [6–14]. The formation of cracks is strongly influenced by the existence of randomly distributed defects that promote cracking. Previous studies of fragmentation of inhomogeneous elastic coatings [15–17] have shown that the average fragment length scales with the applied strain, $\langle L\rangle \propto \varepsilon^{-\kappa}$, the power law exponent depending on the failure threshold distribution and on the *elastic* response to deformation.

Recently, the complex behavior of rocks and solids under deformation has been studied using models containing hysteretic elements [18,19], which ''open'' and ''close'' under pressure. The key hysteretic feature here is that in such elements the thresholds for opening and for closing differ. Such a hysteretic behavior is a nonlinear phenomenon that describes a behavior qualitatively different than elasticity. In particular, the relaxation of stress does not necessarily imply the relaxation of strain (i.e., the plastic behavior appears). Here we study the influence of *plasticity* on fragmentation. For this we extend our former model, by introducing in it plastically deformable elements. Using this extension we investigate both analytically and through simulations the creation and evolution of cracks in the system.

The paper is organized as follows: First we introduce the one-dimensional model mentioned above. Then we determine the equations governing the forces and the elongations in the model. Solving these equations allows us to follow through simulations the extension and the breakage of elements in our model. These results are then discussed and summarized.

We start by noting that the fragmentation of coatings under uniaxial tension often leads to patterns consisting of parallel cracks; then the physical situation is effectively one-dimensional (1D). In line with previous studies of fragmentation [15,20–25] and having such situations in mind, we start from a 1D model, as depicted in Fig. 1. The coating is modeled as a linear arrangement of telescopic elements that can partly extend under tension (for details see below). The number of telescopic elements is $N-1$. Each junction point between two telescopic elements is attached via a leaf spring to the substrate. The leaf springs behave under tension like Hookean springs, i.e., their force-elongation relation is given by $F_k = D v_k$ where F_k (v_k) are the force (the elongation) of the kth leaf spring and D is its elastic constant. Here we only consider the horizontal component of the elongations of the leaf springs and neglect the vector character of the elongation, cf. we follow the shear-lag model [26]. We model the uniform extension of the sub-

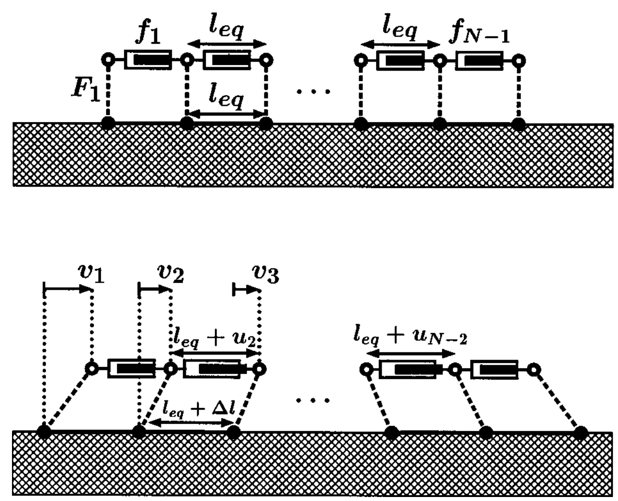

FIG. 1. One-dimensional model for the fragmentation of thin plastic coatings under uniaxial substrate extension. We show the elongations u_k and the forces f_k on the telescopic elements in the surface layer. The elongation of the kth leaf spring is v_k, the corresponding restoring force being F_k. See text for details.

*Present address: ETH Zürich, Department of Materials, Institute of Polymers, 8092 Zürich, Switzerland.

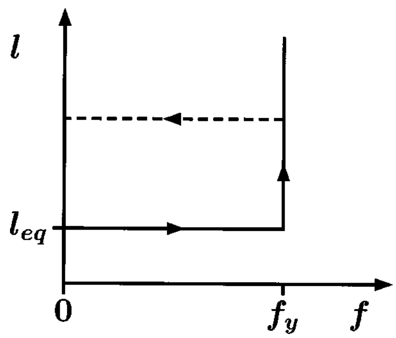

FIG. 2. Force-elongation relation for the telescopic elements. The force is denoted by f and the total length of the element by l. The initial length of the element is l_{eq}. An element starts to open when the force f_k on the element attains the value f_y. The dashed line indicates that the kth telescopic element retains its elongation if the force f_k acting on it again drops below f_y.

strate by increasing the distances between all leaf springs. If the distance between two leaf springs increases from its initial length l_{eq} to $l_{\mathrm{eq}}+\Delta l$, the strain in the substrate is $\varepsilon=\Delta l/l_{\mathrm{eq}}$. Then stress is transferred from the substrate to the coating via the leaf springs. Such a stress transfer results in the elongation of the telescopic elements. Previous studies [15,20–25] of fragmentation of elastic coatings used simple springs instead of telescopic elements.

The force-elongation relation of the telescopic elements is displayed in Fig. 2. The initial length of a telescopic element is l_{eq}. Under (slow) extension of the substrate, the restoring forces f_{k-1} and f_k on the $(k-1)$th and (kth) telescopic elements and the restoring force F_k of the kth leaf spring equilibrate. If the force f_k on the kth telescopic element is smaller than f_y, then its elongation u_k stays at zero: $u_k=0$. If f_k attains the value f_y, the element starts to ''open,'' so that in the following $f_k=f_y$ is obeyed. The elongation takes a determined value $u_k\neq 0$ so that the conditions for the balance of forces and the geometrical requirements of the system of telescopic elements and springs are both fulfilled. We note that such deformations, in which the element elongates at constant force, are quite often observed in the yielding of thermoplastic polymers. Furthermore, as indicated in Fig. 2, if at a later stage the force f_k decreases, the elongation u_k keeps its attained value. The behavior depicted in Fig. 2 models *ideal plasticity*.

We now consider failure in the telescopic elements, say, the mth element breaks if its elongation u_m exceeds a critical value $u_b(m)$. In general, if an element breaks, this leads to stress relaxation in its neighborhood. Hence the stresses of the elements in the vicinity of this new crack decrease. If the force f_k of the kth element drops below the value f_y, then u_k does not decrease to zero, but retains the value that it attained just when the crack occurred. This is indicated by the dashed line in Fig. 2. This value is kept until the force f_k attains again the threshold f_y for opening under the influence of the continuously growing substrate extension. Then the kth telescopic element can again continue to open. Given the stress-strain curve depicted in Fig. 2, our telescopic elements show ''ideal'' plasticity under deformation.

Let us now focus on the breakage of the elements. For this we assume that the $u_b(k)$ discussed above are randomly distributed. Here we take as in former works [5,15,20–23,25] that the cumulative probability distribution $F_{\mathrm{cu}}(u_b)$ of the failure thresholds obeys a power law:

$$F_{\mathrm{cu}}(u_b)=\begin{cases}\left(\dfrac{u_b-u_{\min}}{W}\right)^{\alpha} & \text{for } u_{\min}\leqslant u_b\leqslant u_{\min}+W\\ 0 & \text{otherwise.}\end{cases} \tag{1}$$

The parameters of the distribution are α, W, and $u_{\min}$. As we proceed to show, the values of these parameters have a strong influence on fragmentation.

If the mth telescopic element in the coating breaks, then it is removed from the system irreversibly and does not influence the forces and the elongations anymore, i.e., we set $f_m=0$ and $u_m=0$ after the failure of the mth element.

Our analysis of the model starts with the derivation of the equations for the forces and for the elongations. The geometrical arrangement of Fig. 1 implies

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

for $1\leqslant k\leqslant N-1$. Since the sum of all forces acting on each junction point in the surface layer between two telescopic elements vanishes, we have $F_k=f_k-f_{k-1}$. At the boundaries $f_0=f_N=0$ holds. Hooke's law for the leaf springs implies $F_k=Dv_k$ that leads to $v_k=(f_k-f_{k-1})/D$. Inserting this equation into Eq. (2) yields

$$\Delta l=-\frac{1}{D}(f_{k+1}-2\,f_k+f_{k-1})+u_k. \tag{3}$$

If we divide Eq. (3) by l_{eq} and define $g_k=f_k/(Dl_{\mathrm{eq}})$ and the applied strain $\varepsilon=\Delta l/l_{\mathrm{eq}}$, we are led to

$$\varepsilon=-g_{k+1}+2g_k-g_{k-1}+e_k. \tag{4}$$

In Eq. (4) e_k denotes the relative elongation of a telescopic element: $e_k=u_k/l_{\mathrm{eq}}$.

In the beginning, the elongations both of the telescopic elements and of the leaf springs vanish. Under substrate extension one observes first that only the leaf springs move; at higher extensions also the telescopic elements start to extend. In the first phase, the restoring forces f_k in the surface layer are smaller than f_y, and thus the telescopic elements are not stretched, $e_k=0$. Consequently, we find from Eq. (4)

$$\varepsilon=-g_{k+1}+2g_k-g_{k-1} \tag{5}$$

for $1\leqslant k\leqslant N-1$. The solution of Eqs. (4) with the boundary conditions $g_0=g_N=0$ is a parabola, $g_k=\varepsilon k(N-k)/2$. At larger substrate extension the force on the telescopic element in the middle of the segment attains the value f_y and this element starts to open. This opening prevents a further increase in the force acting on the element. Instead, under further substrate extension the neighboring telescopic elements also open. Then these neighbors also feel the force f_y. Interestingly, it turns out that this process of opening leads to the appearance of a plateau in the e_k vs k plot. Consider exemplarily five consecutive elements, say $n=k-2$, $k-1$, k, $k+1$, $k+2$, for which $f_n=f_y$. With $g_k=f_k/(Dl_{\mathrm{eq}})$ follows from Eq. (4) that the elongations for the elements $m=k-1$, k and $k+1$ are all equal, $e_m=\varepsilon=\Delta l/l_{\mathrm{eq}}$. Hence one observes a successive opening of telescopic elements that show (apart from the flanks) the same elongation; increasing the strain increases the plateau value and also the number of telescopic elements inside the plateau.

In order to exemplify this situation, we have solved the system of Eqs. (4) numerically. In the simulations, we start from our one-dimensional model of Fig. 1. At the beginning, each coating element is assigned a random maximum elongation $u_b(k)$ using Eq. (1). Then the equations for the balance of forces are solved numerically. We applied two different simulation algorithms denoted by *A* and *B*. In our simulations using algorithm *A*, we increase stepwise the applied strain ε starting from $\varepsilon=0$ and solve Eqs. (4) with all e_k equal to zero. If the maximum value of the force f_k exceeds f_y, then the force distribution is determined anew via a relaxation method in order to find which elements open. In our simulations using algorithm *B*, we increase the strain step by step and determine the telescopic element for which f_k exceeds f_y. Then we set its force f_k equal to f_y and determine from Eq. (4) its strain e_k. The use of algorithm *B* requires that the stepwise increment in ε be sufficiently small. Method *A* is slower, but is always successful, whereas method *B* may show instabilities, depending on the situation at hand and hence must be used with care. The e_k obtained by both methods agree within 0.5%.

The results of the computations are presented in Fig. 3 where the parameters are $N=64$, $D=1$, $l_{\mathrm{eq}}=1$, $f_y=1$, $u_{\min}=0.002$, $W=0.01$, and $\alpha=1$. In Fig. 3(a) we plot the relative elongations e_k as a function of k. Figure 3(a) is taken in a situation before the first failure occurs. For the parameters chosen $\varepsilon_y=8g_y/N^2=8f_y/(Dl_{\mathrm{eq}}N^2)=0.001\,95$. One may note the appearance of a plateau, symmetrically located around $k=32$. Figure 3(a) is attained at $\varepsilon=0.003\,75$, so that $\varepsilon>\varepsilon_y$. Figure 3(b) presents through a solid line the forces f_k as a function of k for the same situation as in Fig. 3(a). Note that the force distribution also displays a plateau zone, with $f_k=f_y$. The k range of the plateau zone of the force distribution is identical to that of the strain distribution. Outside the plateau the shape of f_k is parabolic. Additionally, we display in Fig. 3(b) through a dashed line the forces f_k for $\varepsilon=\varepsilon_y$. Then no telescopic element is yet elongated and thus all e_k are zero. Note that for $\varepsilon=\varepsilon_y$ the function f_k vs k is a parabola with its maximal value equal to f_y.

We now turn to the discussion of fracture, where the influence of plasticity becomes important. As just discussed, for $\varepsilon>\varepsilon_y$ the spatial distribution of the strains e_k shows a plateau, with $e_k=\varepsilon$. The telescopic elements at the flanks of the plateau zone are elongated as well, but with $e_k\leqslant\varepsilon$. The elongation of the other elements is zero. Hence only the telescopic elements in the plastic zone (plateau and flanks) can break at this stage. Hence one of the elements of the plastic zone, namely, that with the lowest failure threshold will fail (break) first. In Fig. 3(a) this element is indicated by the star touching the plateau, here at $k=30$. After the failure, the stress in the vicinity of the newly formed crack relaxes, so that next to it the restoring forces f_k of the telescopic elements can drop below f_y. Then the strain e_k of these elements is frozen until (under the influence of the continuous extension of the substrate) at a later stage the force may again attain the value f_y. Due to the randomness in the threshold values, after several breaks the e_k distribution begins to scatter strongly; on the other hand the stress distribution in the sample is less irregular.

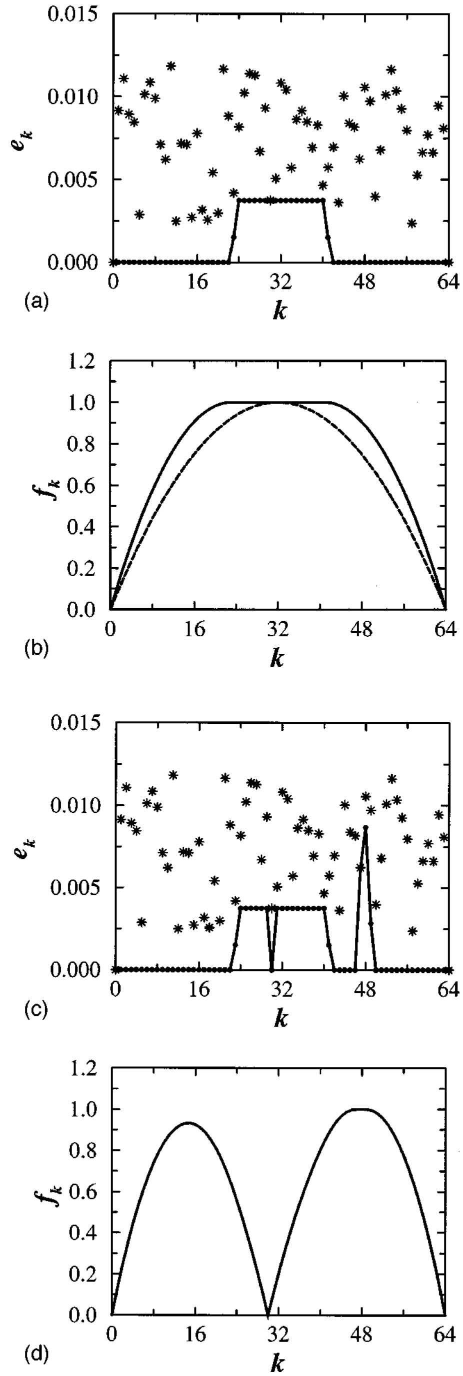

FIG. 3. Evolution of the relative elongations e_k and the force f_k with applied strain ε before the first [(a) and (b)] and the second crack [(c) and (d)] in the surface layer. The solid lines denote the functions f_k and e_k, respectively, the stars are the failure thresholds. The dashed line in (b) is the force distribution for $\varepsilon=\varepsilon_y$. The parameters are $N=64$, $D=1$, $l_{\mathrm{eq}}=1$, $f_y=1$; furthermore $u_{\min}=0.002$, $W=0.01$ and $\alpha=1$ in Eq. (1).

In Fig. 3(c) we show the situation after the first failure occurred (here at position $k=30$), and shortly before the second failure happens (here at $\varepsilon=0.008\,69$). Note that the open elements in the interval $23\leqslant k\leqslant 41$ are now frozen. The force f_k in the two newly formed segments increases with ε. If the maximum value of f_k attains again the value f_y for opening [the plot of f_k vs k is given in Fig. 3(d)], an element in the center of one of the two fragments opens. The second crack occurs in this new plastic zone [here at position $k=47$, see Fig. 3(c)].

As shown in Ref. [15], the evolution of $\langle L\rangle$, the mean crack spacing, under applied strain provides much information about disorder and nonlinear material behavior. Hence here we also monitor the fragmentation kinetics, i.e., the evolution of $\langle L\rangle$ with the applied strain. Here one can distinguish the following three important cases. For large $u_{\min}$, $u_{\min}\gg\varepsilon_y l_{\mathrm{eq}}$, many telescopic elements open before the first failure occurs. Then the strain e_k of almost all coating elements equals ε (''isostrain'' situation [27]). Since the average number of cracks is given by $\langle n\rangle=(N-1)F_{\mathrm{cu}}(\varepsilon l_{\mathrm{eq}})$, the average crack spacing $\langle L\rangle$ is $\langle L\rangle=N/(\langle n\rangle+1)\approx[(\varepsilon l_{\mathrm{eq}}-u_{\min})/W]^{-\alpha}$. We note that this case models an experimentally relevant situation, where cracking always occurs at strains larger than the yield strain.

In the case of large W, $W\gg\varepsilon_y l_{\mathrm{eq}}$, and $u_{\min}$ arbitrary, a few springs may break before large plastic zones form; this affects the initial stage of cracking and lets it resemble the corresponding situation in strongly disordered elastic coatings [5]. On the other hand, for large W, the total number of cracks formed during this stage is small, so that the average crack spacing $\langle L\rangle$ is still large enough to allow for the formation of large plastic zones in later stages of the process. This again will lead to $\langle L\rangle\propto(\varepsilon l_{\mathrm{eq}}-u_{\min})^{-\alpha}$.

Another situation takes place in the case of small W ($W<\varepsilon_y l_{\mathrm{eq}}$) and $u_{\min}<\varepsilon_y l_{\mathrm{eq}}$. In this case only a few elements in the middle of the fragments open. Then the force distribution is nearly parabolic with a maximum value of f_y. Since only a very few elements are open when a crack is formed, each failure occurs when the maximum value of f_k attains f_y. Thus the condition for breakage is $\varepsilon=8g_y/\langle L\rangle^2$, which leads to the power law $\langle L\rangle\propto\varepsilon^{-1/2}$. Here the scaling law exponent is $\frac{1}{2}$, independent of α. In all cases we find a power law $\langle L\rangle\propto\varepsilon^{-\kappa}$, where, however, $\kappa=\alpha$ or $\kappa=\frac{1}{2}$, depending on the parameters.

We have simulated these three different situations: we consider, namely, failure processes in systems given by $D=1$ and $l_{\mathrm{eq}}=1$, while using in the threshold distribution Eq. (1) $\alpha=1$; we consider three cases: in (a) we set $N=256$, $u_{\min}=0.1$, $W=7.5$; in (b) we take $N=64$, $u_{\min}=0$, $W=10$; and in (c) $N=64$, $u_{\min}=0$, $W=0.05$. We note that the set of parameters (a) belongs to the case where $u_{\min}$ is large compared to $\varepsilon_y l_{\mathrm{eq}}$, since we have $u_{\min}=0.1>\varepsilon_y l_{\mathrm{eq}}=0.000\,122$, whereas, evidently, (b) and (c) have $u_{\min}<\varepsilon_y l_{\mathrm{eq}}$. With these parameter sets we perform the simulations for five realizations of the failure threshold distribution and evaluate $\langle L\rangle$ as a function of ε; then we average $\langle L\rangle$ over the five realizations. As stressed above, we expect in the cases (a) and (b) a slope of -1, and for (c) a slope -0.5. The results of the simulations are presented in Fig. 4 together with two lines with slope -0.5 and -1, respectively. As is evident, the simulation data support our analytical expressions.

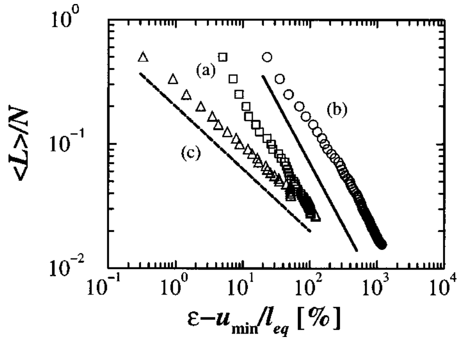

FIG. 4. Average crack spacing $\langle L\rangle$ vs applied strain ε for $D=1$ and $l_{\mathrm{eq}}=1$ and three sets of parameters of the failure threshold distribution Eq. (1). The mean crack spacing for each set of parameters was obtained by averaging over five realizations of the failure threshold distribution. (a) Here $N=256$, $u_{\min}=0.1$, $W=7.5$, $\alpha=1$ and thus $\kappa=1$. (b) The parameters are $N=64$, $u_{\min}=0$, $W=10$, and $\alpha=1$, which yields $\kappa=\alpha$. (c) Here $N=64$, $u_{\min}=0$, $W=0.05$, and $\alpha=1$ holds, and we find $\kappa=1/2$. The slope of the dashed line is -0.5 and the slope of the solid line -1.

Power laws of the type $\langle L\rangle\propto\varepsilon^{-\kappa}$ also show up in the fragmentation of elastic materials; for there one finds in late fragmentation stages $\kappa=\alpha/(2\alpha+1)$ [5,20,21]. We stop to note, however, that in the case of purely elastic materials, the strain inside each separated fragment depends on the actual stress, but is independent of the previous history. Plastic materials, on the other hand, show strains that depend (due to the earlier opening of telescopic elements) on the previous history of the sample. This leads to an additional source of sequential non-Markovian behavior for plastic materials, effect that changes the dependence of κ on α, from the form for elastic materials given above.

We note that the situation discussed above (with purely plastic elements) will persist when the elements also show elastic features at small stresses (note that plasticity dominates at large stresses). The reason is again the formation of large plastic zones at larger strains. Evidently, the initial stages of breakage will depend strongly on the elastic parameters involved [5].

Summarizing, in this study we have investigated the sequential cracking of thin coatings under uniaxial substrate extension. We have put forward a 1D model with ideal plastic elements. This allowed us to focus on the interplay between the plastic behavior of the coating and the random distribution of coating defects. Our numerical simulations based on this model reveal that the values of the parameters entering Eq. (1) (the failure probability distribution) strongly influence the shape of the strain distribution, since they determine the size of the plastic zone. The fragmentation kinetics (as determined from simulations) obeys scaling, i.e., $\langle L\rangle$ and ε are related by a power law behavior.

The authors are thankful for the support of the DFG (via SFB 428) and of the Fonds der Chemischen Industrie.

[1] *Disorder and Fracture*, Vol. 235 of *NATO Advanced Study Institute, Series B: Physics*, edited by J. C. Charmet, S. Roux, and E. Guyon (Plenum, New York, 1990).
[2] *Statistical Models for the Fracture of Disordered Media*, edited by H. J. Herrmann and S. Roux (North-Holland, Amsterdam, 1990).
[3] B. K. Chakrabarti and L. G. Benguigui, *Statistical Physics of Fracture and Breakdown in Disordered Systems* (Clarendon, Oxford, 1997).
[4] J. A. Nairn and S.-R. Kim, Eng. Fract. Mech. **42**, 195 (1992).
[5] U. A. Handge, Y. Leterrier, G. Rochat, I. M. Sokolov, and A. Blumen, Phys. Rev. E **62**, 7807 (2000).
[6] M. S. Hu and A. G. Evans, Acta Metall. **37**, 917 (1989).
[7] J. W. Hutchinson and Z. Suo, Adv. Appl. Mech. **29**, 63 (1992).
[8] D. R. Wheeler and H. Osaki, in *Metallization of Polymers*, edited by E. Sacher, J.-J. Pireaux, and S. P. Kowalczyk, ACS Symposium Series 440 (ACS, Washington D.C., 1990), p. 500.
[9] B. Ollivier, S. J. Dowey, S. J. Young, and A. Matthews, J. Adhes. Sci. Technol. **9**, 769 (1995).
[10] W. A. Curtin, J. Mater. Sci. **26**, 5239 (1991).
[11] J. C. Grosskreutz and M. B. McNeil, J. Appl. Phys. **40**, 355 (1969).
[12] H. Colina, L. de Arcangelis, and S. Roux, Phys. Rev. B **48**, 3666 (1993).
[13] A. Mézin and B. Sajid, Thin Solid Films **358**, 46 (2000).
[14] C.-Y. Hui, S. L. Phoenix, M. Ibnabdeljalil, and R. L. Smith, J. Mech. Phys. Solids **43**, 1551 (1995).
[15] U. A. Handge, I. M. Sokolov, and A. Blumen, Phys. Rev. E **61**, 3216 (2000).
[16] J. Andersons, U. A. Handge, I. M. Sokolov, and A. Blumen, Eur. Phys. J. B **17**, 216 (2000).
[17] U. A. Handge, I. M. Sokolov, A. Blumen, E. Kolb, and E. Clément, J. Macromol. Sci., Phys. **B38**, 971 (1999).
[18] R. A. Guyer, K. R. McCall, and G. N. Boitnott, Phys. Rev. Lett. **74**, 3491 (1995).
[19] R. A. Guyer and P. A. Johnson, Phys. Today **52** (4), 30 (1999).
[20] O. Morgenstern, I. M. Sokolov, and A. Blumen, Europhys. Lett. **22**, 487 (1993).
[21] O. Morgenstern, I. M. Sokolov, and A. Blumen, J. Phys. A **26**, 4521 (1993).
[22] T. Hornig, I. M. Sokolov, and A. Blumen, Phys. Rev. E **54**, 4293 (1996).
[23] I. M. Sokolov and A. Blumen, Physica A **266**, 299 (1999).
[24] U. A. Handge, I. M. Sokolov, and A. Blumen, Europhys. Lett. **40**, 275 (1997).
[25] U. A. Handge, Y. Leterrier, J.-A. E. Månson, I. M. Sokolov, and A. Blumen, Europhys. Lett. **48**, 280 (1999).
[26] H. L. Cox, Br. J. Appl. Phys. **3**, 72 (1952).
[27] H. D. Wagner, O. Lourie, Y. Feldman, and R. Tenne, Appl. Phys. Lett. **72**, 188 (1998).