Energy landscape view of fracture and avalanches in disordered materials

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Molecular simulations are carried out to probe how strain-induced changes in the energy landscape are related to fracture processes in disordered systems. The simulations address a two-dimensional system that consists of 9952 particles with a distribution of sizes, and the changes in the structure and properties with strain are determined with the system constrained to an energy minimum. As the system is strained, local minima of the energy landscape are found to flatten out and disappear, which causes discontinuous structural rearrangements. These structural rearrangements, which correspond to avalanche events, lead to void nucleation and crack growth in discrete steps.

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I. INTRODUCTION

Glassy materials are often used in applications where their response to stress is important. For example, silica-based glasses and polymethylmethacrylates are used for windows, polystyrenes for bicycle helmets, polycarbonates for bullet-proof glass, and epoxy resins in aircraft structural components. In these applications, the tendency of the material to stress-induced fracture is an important consideration.

Most previous molecular simulations addressing fracture have focused on crystalline systems [1–4], where the mechanism of crack propagation is closely related to dislocation defects. Since dislocations are not relevant in disordered systems, the mechanism of crack propagation in disordered systems will differ from that in crystalline systems. A recent molecular dynamics study shows that crack propagation in disordered systems occurs through two-state transformations in localized transformation zones [5,6]. Also, phenomenological models have shown that the approach to fracture in disordered systems is marked by discrete avalanchelike events [7–10]. The present paper investigates the strain-induced changes in the potential energy landscape that underlie crack propagation in disordered systems, and these changes in the landscape are related to localized transformation zones and avalanche events.

II. COMPUTATIONAL METHODS

Simulations are carried out for a two-dimensional system with 9952 particles. A two-dimensional system is used rather than a three-dimensional system because finite size effects are smaller (for a given number of atoms), and because crack growth is more easily visualized. Periodic boundary conditions are used in the direction that the system is strained, but not in the other direction—the omission of periodic boundary conditions in this direction allows greater distortion of the system under strain. The particles interact through the Weber and Stillinger potential [11]

$$V(r)=\begin{cases} \varepsilon A\left[\left(\dfrac{r}{\sigma_{ij}}\right)^{-p}-\left(\dfrac{r}{\sigma_{ij}}\right)^{-q}\right]e^{-[1/(a-r/\sigma_{ij})]} & (r/\sigma_{ij}<a) \\ 0 & (r/\sigma_{ij}\geqslant a) \end{cases}, \tag{1}$$

where $p=12$, $q=0$, $a=1.652\,194$, $A=8.805\,997$, and $\sigma_{ij}=(1/2)(\sigma_i+\sigma_j)$; the parameter ϵ scales the well depth, and σ_i describes the size of the particle i. This potential is similar to the Lennard-Jones potential, but has the advantage of a finite interaction distance with all derivatives continuous at the interaction cutoff distance. The continuity of all derivatives is important in this investigation because changes in the normal mode frequencies with strain are examined; if the second derivatives were discontinuous at the cutoff distance, artificial discontinuous changes in the normal mode frequencies with strain would occur whenever interactions move into or out of the cutoff distance.

A distribution of particle sizes (σ_i) is used to cause the system to be disordered, since two-dimensional systems with identical particles do not quench to fully disordered structures at low temperature. The particle sizes are chosen from a Gaussian distribution with a mean value of σ_0 and a standard deviation of 20%. Initially, the system consists of 10 000 particles in an ordered hexagonal structure at the density $\rho=(2/\sqrt{3})\sigma_0^{-2}$. The particles are then allowed to relax to the minimum energy configuration (which is disordered due to the distribution of particle sizes), at constant density. Particles are then removed to form an initial crack in the system, such that 9952 particles remain in the system. After the crack is formed, the particles are again allowed to relax to the minimum energy configuration at constant density.

The simulation method determines how the structure and properties associated with a single energy minimum change

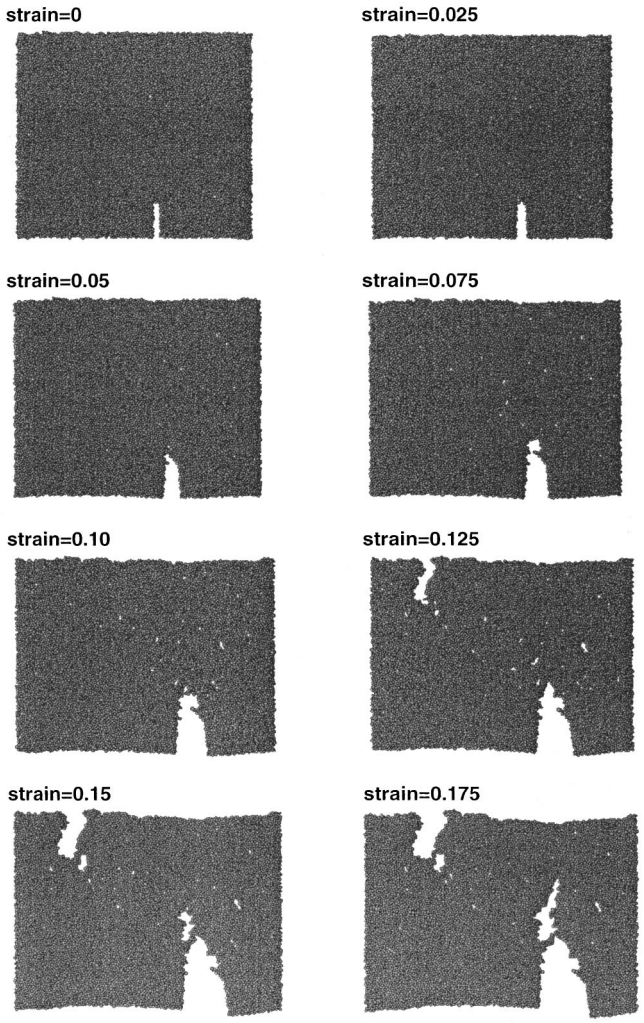

FIG. 1. Structure of material as a function of strain.

with strain. The simulation proceeds by increasing one dimension of the simulation cell in extremely small strain increments that approximate continuous change (tensile strain increments of 0.0001 are used). After each strain increment, the potential energy is minimized with respect to the atomic coordinates (at constant strain), and the stress is evaluated at the energy minimum. These simulations are also carried out on a smaller system (790 particles), for which the normal mode eigenvalues and eigenvectors are also calculated at the energy minima.

III. RESULTS

The changes in structure with strain are shown in Fig. 1. No significant structural changes occur until the strain exceeds 0.025, and the initial crack grows to about twice its original length by a strain of 0.125. Voids also form in the material at high strains. A second crack forms on the other side of the material at a strain of 0.121.

The results for the tensile stress as a function of strain are shown in Fig. 2. A linear (elastic) regime exists at low strains (<0.025), and a nonlinear (inelastic) regime exists for higher strains (>0.025). Note that the onset of the nonlinear regime

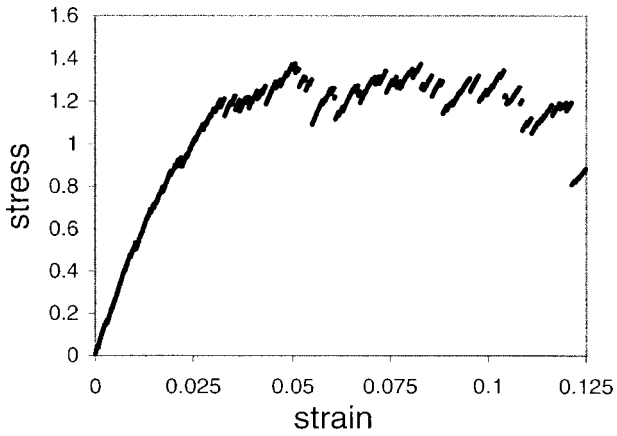

FIG. 2. Tensile stress as a function of tensile strain.

coincides with the start of crack growth and void nucleation (shown in Fig. 1). In the nonlinear regime, stress usually increases continuously and nearly linearly with strain, but these increases are punctuated by discontinuous stress drops. Such discontinuous stress drops have been observed in previous simulations of strained glasses [12–15].

Insights regarding fracture are obtained by a more detailed comparison of the structural results to the stress-strain results. As shown in Fig. 3, void nucleation and crack growth occur through discrete and discontinuous structural changes that coincide with the discontinuous stress drops, and the structure does not change significantly between these discontinuous changes. The crack growth mechanism is seen to begin with the nucleation of a void near the crack tip [Fig. 3(bi)], where local stresses are greatest. The void then grows in discrete steps [Figs. 3(bii) and 3(biii)], and ultimately coalesces with the crack [Fig. 3(biv)]. This crack growth mechanism is found for all instances of crack growth in the present simulations (results not shown for the other instances), and this mechanism is similar to that found previously in simulations of a glassy Lennard-Jones system [5]. The mechanism by which the new crack forms consists of the nucleation and growth of several voids [Figs. 4(bi)–4(biii)], followed by the coalescence of these voids with each other and with the surface of the material [Fig. 4(biv)]. We emphasize again that this mechanism proceeds in discrete steps that punctuate periods of little structural change.

To understand the origin of the discontinuous structural changes associated with crack growth, the strain dependences of the normal mode eigenvalues are examined for a 790-particle system (the normal mode calculation is not feasible for the 9952-particle system). As shown in Fig. 5, the discontinuous structural change associated with crack growth, and the accompanying discontinuous stress drop, occur when the lowest normal mode eigenvalue of the system decreases to zero. The normal mode eigenvalues correspond to the curvature of the energy surface in orthogonal directions, and the decrease of a normal mode eigenvalue to zero implies the flattening out and disappearance of a local energy minimum, as shown schematically in Fig. 6. After the local minimum disappears, the system becomes mechanically unstable (i.e., there are nonzero net forces on the particles), and the system undergoes a structural rearrangement to an alter-

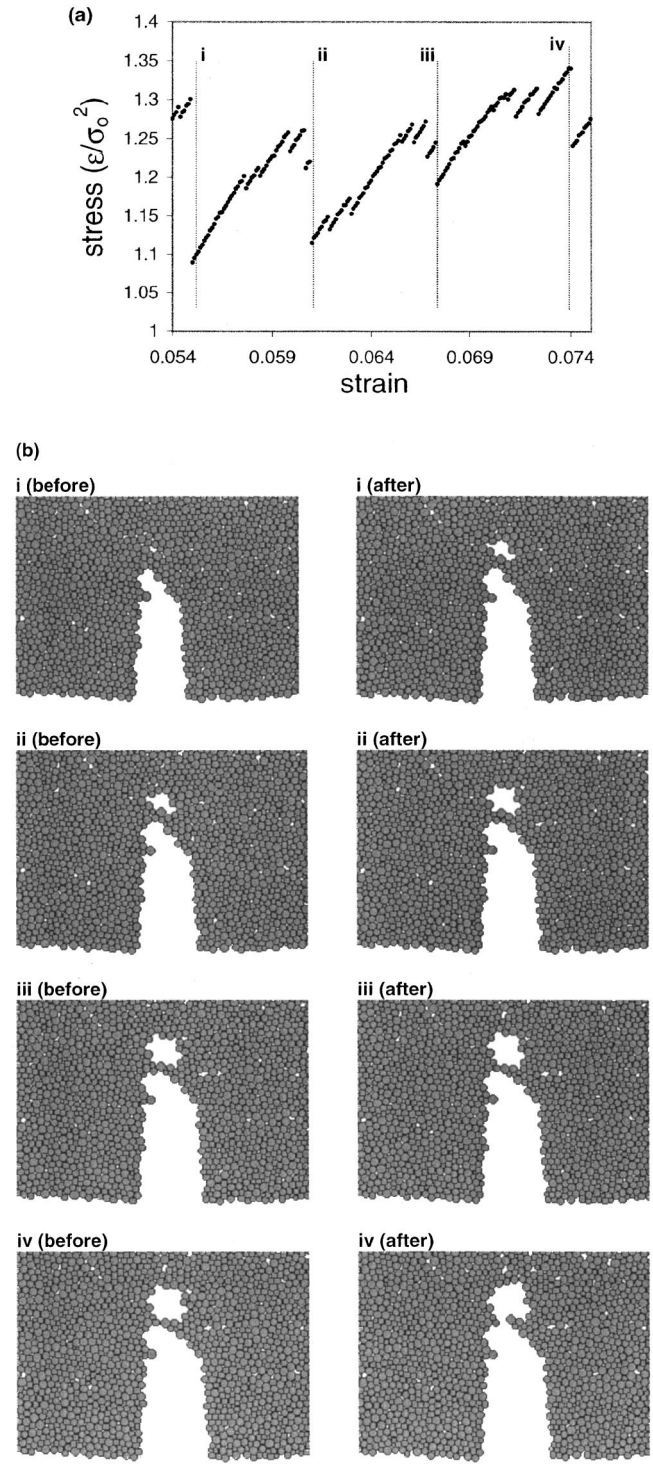

FIG. 3. Detailed description of the discontinuous changes that lead to crack growth. (a) Stress vs strain. (b) Structures before and after the discontinuous changes at strains denoted in (a).

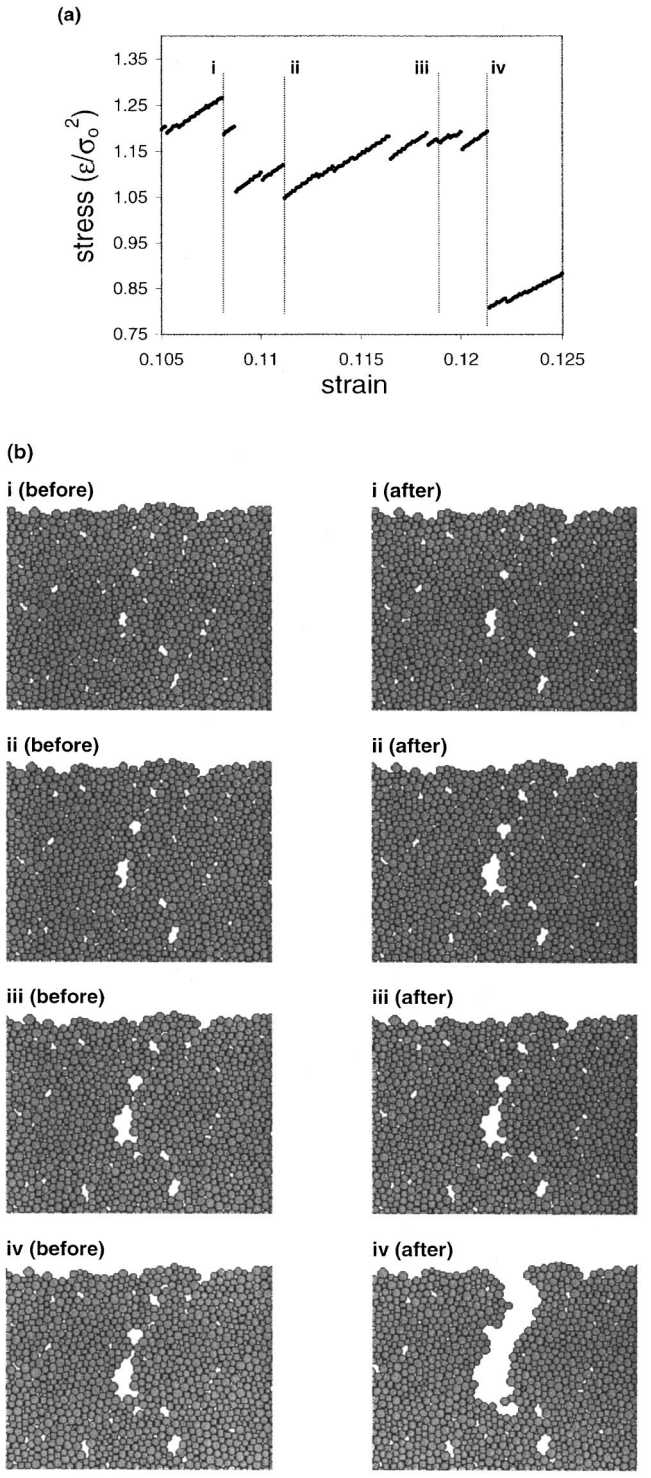

FIG. 4. Detailed description of the discontinuous changes that lead to crack formation. (a) Stress vs strain. (b) Structures before and after the discontinuous changes at the strains denoted in (a).

nate potential energy minimum. The structural rearrangements can be predicted before they occur by an analysis of the normal mode eigenvector associated with the soft mode: As shown by the insets in Fig. 5, the particles that move most in the structural rearrangement correspond to the particles with the largest components of this eigenvector. These strain-induced disappearances of energy minima in disordered systems have been examined in more detail elsewhere, by following the strain-induced changes in the height, position, and curvature of the barrier along the reaction coordinate [16,17] The present results show that void nucleation, void growth, and void-crack coalescence occur in discrete steps associated with these disappearances of energy minima.

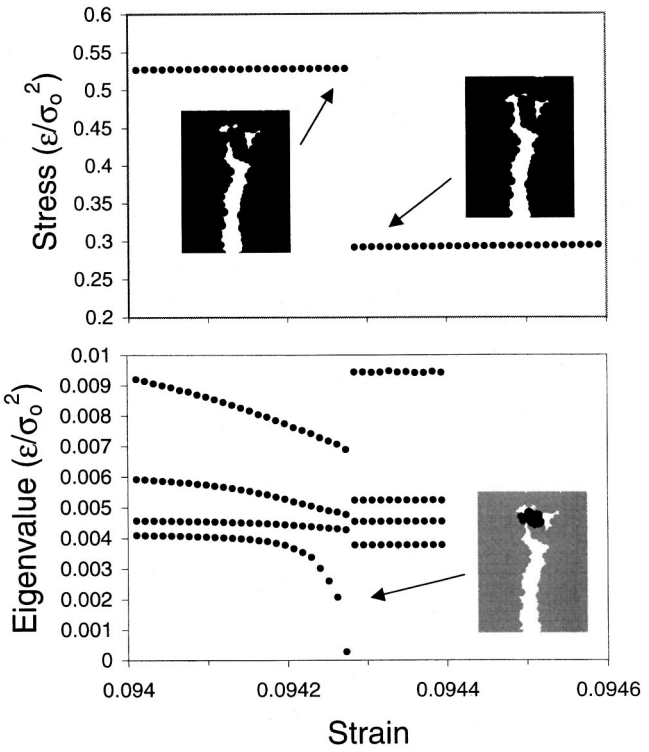

FIG. 5. Stress and the lowest nonzero normal mode eigenvalues vs strain. The structures before and after the discontinuous change in stress are shown as insets in the stress plot, and the ten particles with the largest components of the eigenvector corresponding to the soft mode eigenvalue are shown in the inset in the eigenvalue plot.

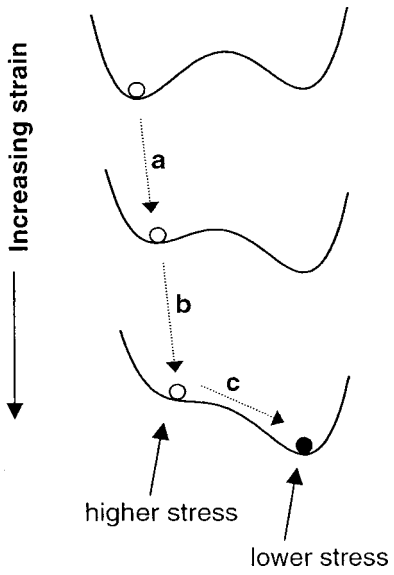

FIG. 6. Schematic representation of a strain-induced disappearance of an energy minimum. The curves represent the potential energy along the relevant coordinate, and the circles represent the state of the system. The change in the state of the system denoted by arrow ''c'' corresponds to the structural rearrangement following the disappearance of an energy minimum.

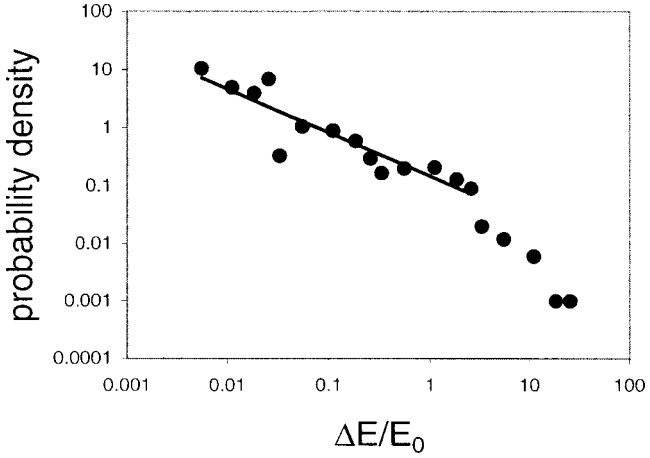

FIG. 7. Probability distribution for avalanche events releasing energy $\Delta E/E_0$, where E_0 is the average binding energy per particle.

The structural rearrangements following the disappearances of energy minima correspond to avalanche events, and the size of these events can be characterized by the energy released ΔE [18]. The probability density for events releasing energy ΔE is shown in Fig. 7. The smaller-scale structural rearrangements are more probable, and follow a power law distribution with an exponent of -0.81 [i.e., Prob$(\Delta E) \sim \Delta E^{-0.81}$]. The larger-scale rearrangements do not follow a power law distribution, but decrease exponentially in probability. These results are very similar to simulation results for a sheared foam based on a model much like the one used here [19]; the avalanche events observed in these foam simulations likely arise from the strain-induced disappearance of energy minima described here. The lack of power law behavior observed here at larger scales may be an artifact due to finite-system-size effects [8,9], or it may be a real effect in that the dissipation of all energy in these zero-temperature simulations may inhibit large-scale structural rearrangements [19,20]—further work is necessary to resolve this issue.

The number of particles involved in the structural rearrangements following the disappearance of energy minima can be estimated by the participation number $p = \Sigma_{i=1}^{N} (d_i/d_{\max})^2$, where d_i is the displacement of particle i (with respect to affine motion), $d_{\max}$ is the largest displacement undergone by any one particle, and N is the number of particles [21]; $p=1$ if only one particle undergoes a displacement, and $p=N$ if all particles undergo equal displacements. Figure 8 compares the probability distribution of the participation number in the case in which the energy minimum is stable during a strain increment to that in the case in which the energy minimum disappears during a strain increment. The frequency distribution is peaked at $p \sim 3000$ when the energy minimum is stable, and at $p \sim 30$ when the energy minimum disappears. These results indicate that the particle displacements following strain-induced disappearance of energy minima are localized to $\sim$10–100 particles [22]. Furthermore, there is no systematic particle-size dependence of the participation in avalanche events.

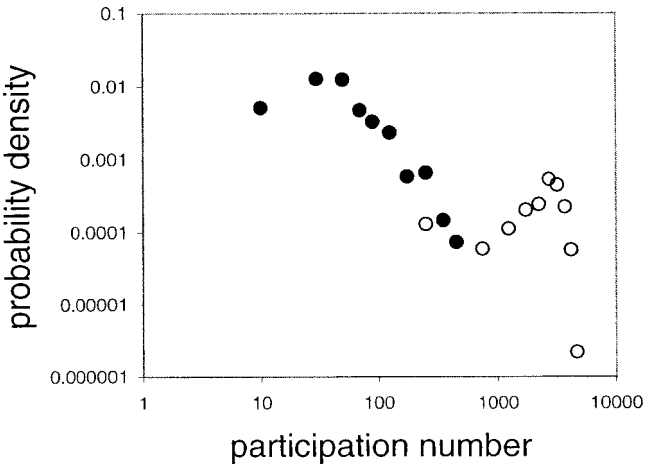

FIG. 8. Probability distribution for the participation numbers for structural changes after strain increments. Filled symbols, strain increments involving disappearances of energy minima; open symbols, strain increments where energy minima remain stable.

IV. DISCUSSION

The present results show how crack growth and avalanche events are related to the changes in the energy landscape with strain. The structural changes observed here apply to real systems in the limit of low temperature, for which the system always remains at energy minima; higher temperatures can cause the system to follow other paths over the energy landscape. The applicability of the present results to real systems also requires that the strain rate be low in comparison to the rate of relaxation within an energy minimum, so that the system could remain near a local minimum as it is strained (note that the rate of relaxation within an energy minimum is extremely high—on the order of the vibrational frequencies).

The structural changes following the disappearance of energy minima correspond to the two-state transformations in localized transformation zones discussed previously in regard to crack propagation [5,6]. The previous molecular dynamics investigation identified the transformation zones after the fact, by observing actual transformations, and the question was raised of how to identify the zones, at least in principle, before the transformation occurs [6]. The present results provide an answer to this question: An incipient transformation is indicated by the decrease of a normal mode eigenvalue to zero, and the particles involved in the transformation can be identified before the transformation occurs from the eigenvector corresponding to this soft mode eigenvalue.

It has been suggested that the fracture process occurs when stress (or strain) brings the material past its spinodal [23]. A spinodal represents the limits for which a phase can exist, even as a thermodynamically metastable state, because the phase is mechanically unstable past the spinodal. The present results show that strain causes the energy minima corresponding to the unfractured material to disappear. The present process is analogous to crossing the spinodal of the material, as the unfractured material cannot exist even as a metastable state after these energy minima disappear; however, this process takes place over a broad strain range due to the variety of local environments that are ''frozen'' into a glassy system (i.e., different local particle arrangements become unstable at different strains), whereas a thermodynamic spinodal occurs at a particular strain.

The present results relating void formation to the strain-induced changes of *individual* energy minima complement previous results that relate void formation to strain-induced changes of the *average* characteristics of energy minima [24]. The present result that tensile stress increases with increasing strain at low strains where voids do not exist, but decreases with increasing strain at high strains after voids appear, is analogous to the previous result that pressure decreases with increasing volume at small volumes where voids do not exist, but increases with increasing volume at large volumes after voids appear (note that pressure corresponds to a negative isotropic tensile stress). The present focus on the strain dependence of single energy minima elucidates the mechanism of void formation in the energy landscape as being due to disappearances of energy minima of the homogeneous material.

The strain-induced disappearances of energy minima that give rise to crack growth in discrete steps arise independently of the initial crack inserted in the material, and even occur in the absence of an initial crack. The strain-induced disappearances of energy minima also give rise to other phenomena in disordered materials under stress, including shear-induced diffusion [25], amorphous-amorphous phase transitions [26,27], and the reversal of aging in glasses [15].

V. CONCLUSIONS

Molecular simulations show that the energy landscape of a disordered material is deformed as the material is strained, such that local minima flatten out and disappear. If the system is at a local minimum that disappears, the system will undergo a sudden structural rearrangement to an alternative local minimum. These sudden structural rearrangements correspond to avalanche events. The size distribution of the avalanche events follows a power law behavior for smaller avalanche sizes, and an exponential decrease for larger avalanche sizes (the exponential decrease may be a finite-size effect), where the size of an event is characterized by the energy released.

The fracture process proceeds (in these zero-temperature simulations) in discrete steps that correspond to the structural rearrangements following the disappearances of energy minima. Crack propagation occurs by the nucleation of a void near the crack tip, followed by the growth of the void, and then the coalescence of the void with the crack to yield a longer crack.

ACKNOWLEDGMENT

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