# Early-time instabilities in a dynamic percolation model

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A simple site-bond cluster dynamics model is proposed in which clusters evolve by a combination of single monomer aggregation, merger, and dissociation processes. The evolution of clusters after a quench from the stable sol phase to the "unstable" gel phase is followed by Monte Carlo simulation. Comparisons are made to the evolution of clusters in the very early stages of spinodal decomposition.

## I. INTRODUCTION

The application of cluster-growth models and the ideas of connectivity have led to much progress in our understanding of the temporal evolution of a variety of geometrical[1] and thermal[2] phenomena. Applications to thermal systems have included critical phenomena,[3] nucleation,[4,5] and spinodal decomposition.[6] However, the application of connectivity and other geometrical considerations to thermal problems is made difficult by the problem of obtaining the appropriate definition of connectivity. In order to avoid this difficulty and to gain insight into the nature of the very early stages of spinodal decomposition, we develop in this paper a simple cluster dynamics model for which the definition of connectivity is clear. We then use Monte Carlo methods to investigate the spinodal-like instability in this model.

If there is any utility to relating thermal problems such as nucleation and spinodal decomposition to connectivity problems, there must be an intrinsic spinodal-like instability in the connectivity problem. Some evidence that such an instability does occur in connectivity problems was obtained recently in a mean-field treatment of random continuum percolation.[7] Solutions for the order parameter and the mean cluster size (the analog of the susceptibility in thermal systems) were obtained which were similar to mean-field solutions of spinodal unstable regions in thermal systems. In particular, a solution exists for a range of parameters which corresponds to an unstable phase characterized by a negative mean cluster size and an order parameter with zero magnitude.[8]

As an example we first discuss the application of the ideas of connectivity in the context of nucleation in a lattice-gas model. Near the coexistance curve and far from the critical point (see Fig. 1), it is natural[4] to choose the droplets to be site percolation clusters in which occupied nearest-neighbor sites are connected; the probability of an occupied site is given by the usual lattice-gas Boltzmann factor. This definition of droplets is useful near the coexistence curve but breaks down either for deep quenches or near the critical point.[3] As the quench depth is increased, the nearest-neighbor droplets reach the percolation threshold before the limit of metastability is reached. That is, this definition of droplets implies that an infinite droplet exists in a system which has not yet nucleated. This problem has been removed by proper definitions of droplets.[5]

The successful application of cluster ideas and the appropriate choice of connectivity is more difficult for spinodal decomposition.[4–6] The absence of a quasiequilibrium theory such as that which exists in the nucleation problem, implies that the method of Ref. 5 cannot be used directly to select the correct clusters for spinodal decomposition.

We take "correct" clusters to mean that the pair-connectedness function (the probability that two occupied sites a distance $r$ apart are in the same cluster) is isomorphic to the pair correlation function (which is related to the probability that two occupied sites are a distance $r$ apart). The most careful definition of clusters for deep quenches can be obtained in the vicinity of the mean-field spinodal,[5,9] where the appropriate definition of the drop-

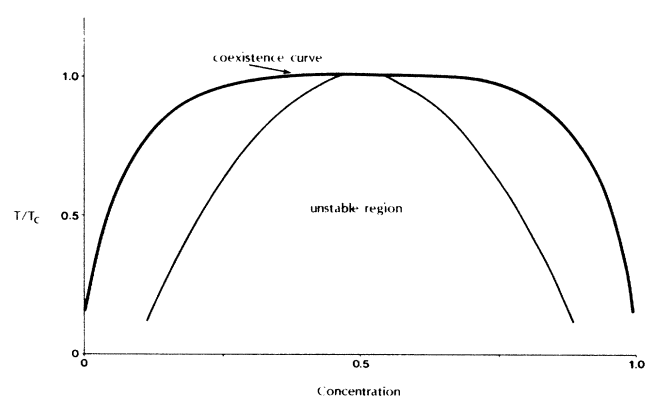


FIG. 1. The coexistence curve and limit of metastability (inner curve) for the Ising model. The simple picture of nearest-neighbor droplets breaks down either for deep quenches or quenches near the critical point.

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lets is given by correlated site-bond percolation clusters. The advantages obtained by the above definition of connectivity are considerable. For example in the nucleation problem, the correct definition of nucleation droplets allows the measurement by computer simulation of nucleation rates and growth characteristics and has led to a theory of crossover from deep to shallow quench nucleation for long-range interactions.[10] The concepts and techniques of cluster-growth methods applied to the spinodal decomposition problem could also yield detailed predictions for the early-time behavior which might be observable in computer simulations. Crossover from one time regime to another could be followed as the evolution of well-defined connectivity mechanisms. Another benefit would be that the reverse mapping would allow us to apply field-theoretic formulations[11,12] and renormalization-group methods,[13,14] already successfully applied to spinodal decomposition problems, to cluster-growth models which describe the kinetics of gelation.

We introduce our site-bond cluster dynamics model in Sec. II A; a "dynamics" is chosen which allows the clusters to dissociate and merge with time. The Monte Carlo simulations of the model in two dimensions are described in Sec. III. One of the main results of the simulations is that after a quench from the sol phase to the gel phase, the evolution of the largest cluster proceeds in two stages corresponding to single monomer aggregation followed by the merger of clusters of comparable size. In Sec. IV we discuss the implication of our results for the interpretation of the early stages of spinodal decomposition.[7,11,12]

## II. DEFINITION OF THE MODEL

We adopt a site-bond cluster dynamics model in which a combination of single monomer growth, merger, and dissociation of clusters can occur. A cluster is defined as a set of occupied sites connected by occupied bonds. Perimeter bonds are unoccupied bonds which are connected to two occupied sites one of which is in a cluster. At time $t=0$, sites and bonds are occupied at random with initial probabilities $p_s^0$ and $p_b^0$, respectively. The system is then "quenched" by reoccupying all lattice sites with a new probability $p_s$. Occupied bonds which are not between two newly occupied sites are culled. At $t=1$ the clusters grow by occupying all perimeter bonds with probability $p_b$; during this time clusters grow or merge. After one time step, all perimeter bonds have been tested once for occupancy. At $t=2$ we reoccupy all lattice sites with probability $p_s$ and bonds which are no longer connected are removed. The clusters are then redefined in terms of the newly occupied sites and the remaining bonds, and the new perimeter bonds are obtained. The perimeter bonds are then "grown" with probability $p_b$.

The above procedure is continued until the cluster-mass distribution, $n_m(t)$, and the average mass of the clusters becomes independent of time. The radius of gyration $R$ of a cluster is defined in the usual way as

$$R^2=\left[\sum_{i=1}^{m}(r_i-r_{\text{c.m.}})^2\right]\Big/m \quad ,$$

where $r_i$ is the position at time step $t$ of the $i$th occupied site in the cluster, $r_{\text{c.m.}}$ is the center of mass of the cluster, and the mass $m$ of a cluster is the number of occupied sites of the cluster.

Note that our site-bond cluster dynamics model differs from ordinary percolation. In the latter, configurations are produced by randomly occupying all sites with probability $p_s$ and each bond connected to two occupied sites with probability $p_b$. At each time step in our dynamical model all sites are occupied with probability $p_s$ and bonds from the previous time step which no longer have two occupied sites are removed. There are two competing dynamical effects. The reoccupancy of sites after every time step and the culling of "excess" bonds leads to a concentration of bonds which might be less than $p_b$. On the other hand the "growth" of bonds at every time step implies that a given bond may be tested for occupancy several times. Hence after a long time the concentration of bonds might be greater than $p_b$. The static universality class of the (long-time) equilibrium properties of the model is not known, although it is reasonable to assume that the model is in the same universality class as ordinary percolation.

## III. RESULTS

The Monte Carlo simulations were performed on a $128\times128$ square lattice with periodic boundary conditions; for comparison some simulations were also performed on $64\times64$ and $256\times256$ square lattices. Many of the runs were performed with only one trial; in other cases averages were taken over ten trials. No significant differences between the behavior for one trial and that averaged over ten trials were found.

### A. Phase diagram

The gel phase is characterized by the presence of a spanning cluster in contrast to the sol phase which is characterized by finite-sized clusters. Since we adopt periodic boundary conditions, the most useful definition of a spanning cluster is not obvious. We operationally define the critical surface separating the sol and gel phase by two features of the cluster-distribution data. For values of $p_s$ and $p_b$ which are clearly in the gel phase, it is observed that the largest cluster has a radius of gyration $R$ on the order of 26 for the $128\times128$ lattice. The existence of such a cluster is also associated with the observation that the cluster-mass distribution is characterized by a lack of intermediate clusters of mass and radius of gyration between the largest cluster and the many small clusters. These considerations lead us to conclude that the gel phase is characterized by the presence of a cluster with $R$ on the order of 26 and no clusters of intermediate mass and length. The values of $p_s$ and $p_b$, which separate the region where these two features are present from the region where they are both absent, is taken to be the critical surface.

We show the phase diagram for the site-bond cluster dynamics model in Fig. 2. For comparison we show the results for the phase diagram for site-bond percolation as obtained in Ref. 15 using simple renormalization-group arguments for small cells. It is important to remember

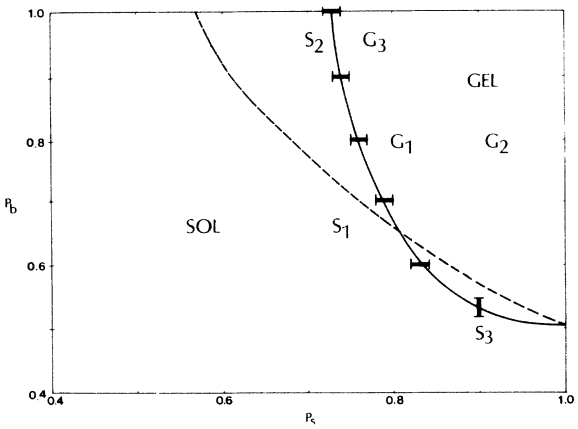


FIG. 2. Critical surface for the site-bond cluster dynamics model. For comparison the critical surface (from Ref. 15) for ordinary site-bond percolation is also shown. The gel phase is characterized by a spanning cluster and many small clusters; the sol phase is characterized by many finite-sized clusters. The particular values of $p_s$ and $p_b$ discussed in the text correspond to $S_1=(0.7,0.75)$, $S_2=(0.7,0.95)$, $S_3=(0.9,0.5)$, $G_1=(0.8,0.8)$, $G_2=(0.9,0.8)$, and $G_3=(0.75,0.95)$.

that the parameters $p_s$ and $p_b$ in our model are dynamic quantities and are not the same as the site and bond probabilities as they are defined in the usual site-bond percolation model. For large values of the parameter $p_b$, the observed concentration of occupied bonds found in the long-time limit is less than $p_b$, and therefore the critical surface is shifted to the right. On the other hand, for $p_b$ approximately equal to 0.5, the concentration of occupied bonds is greater than $p_b$ and the critical surface is shifted to the left.

### B. Stages of cluster growth

The probabilities associated with the initial configuration are taken to be in the sol phase. The corresponding initial configurations typically have a uniform density of small clusters with an average mass of two occupied sites. In general, the long-time behavior is independent of the site and bond probabilites which are used to obtain the initial cluster distribution. However, in the vicinity of $S_2$ and $S_3$ (see Fig. 2) the initial configuration must be carefully prepared to avoid finite-size effects.

The competition between site reoccupancy, bond removal, and bond growth determines the evolution of the clusters. We expect that the qualitative features of the evolution of the clusters to be independent of the details of the "dynamics" of the model. A qualitative understanding of the nature of the cluster dynamics can be obtained from the time dependence of the cluster-mass distributions shown in Figs. 3(a)—3(c) and the "snapshots" of the configurations shown in Figs. 4—6.

We first discuss the behavior of the system as we quench from deep in the sol phase to a region in the sol phase near the critical surface. For $p_s=0.75$ and $p_b=0.7$ ($S_1$ in Fig. 2), we see from the cluster-mass distribution shown in Fig. 3(a) that the number of clusters increases but the average mass of the clusters remains roughly constant during the first few time steps. After a long time ($t=40$), the cluster distribution reaches a steady state and the peak is at a mass approximately equal to 10. For quenches closer to the critical surface, the cluster-mass distribution broadens and the peak shifts to larger values. The time necessary to come to a steady state also increases, indicating the existence of critical slowing down.

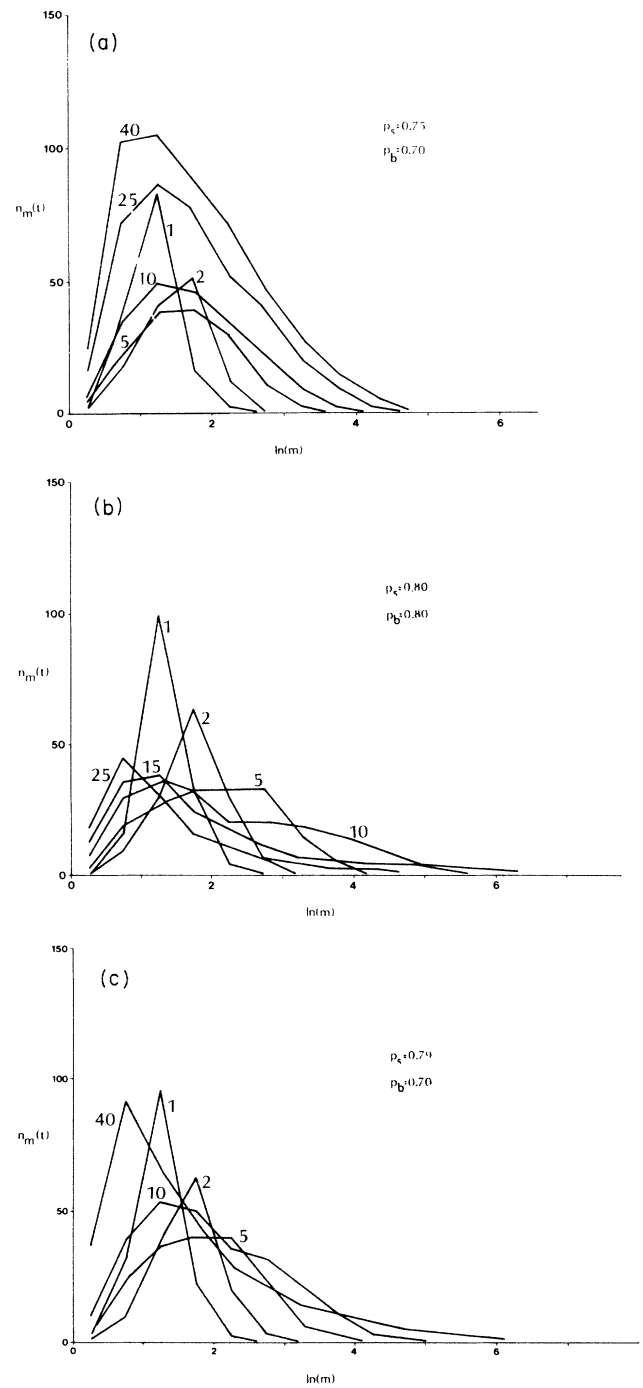


FIG. 3. The cluster mass distribution $n_m$ as a function of the log of the mass $m$. The numbers given denote the number of time steps. (a) corresponds to the sol phase; (b) corresponds to the gel phase; (c) corresponds to the critical surface.

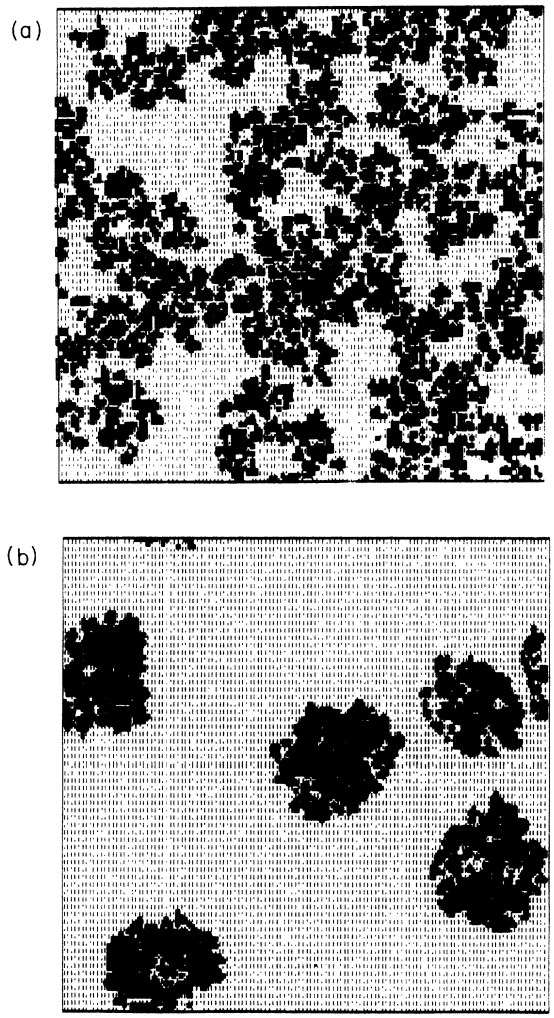


FIG. 4. Snapshots of configurations after 30 time steps for the system quenched into sol phase. (a) corresponds to $S_1$, where $p_s=0.7$ and $p_b=0.75$, and (b) corresponds to $S_3$, where $p_s=0.9$ and $p_b=0.5$.

There are two dominant types of dynamical behavior for quenches in the sol region. For $p_s$ near 0.5 (near $S_1$ and $S_2$), the dominant behavior is characterized by small isolated clusters which do not grow large enough to merge [see Fig. 4(a)]. If large clusters are created artificially in this region, the clusters disassociate into many small clusters. Since at each time step the probability that a bond is removed is $1-p_s^2$, many bonds are broken when $p_s$ is small and the growth of the clusters is inhibited. At the other end of the sol region, e.g., $S_3$, the clusters become larger and break off into more than one cluster to form neighboring disconnected groups of clusters. Although the number of clusters has increased, the group of disconnected clusters does not grow large enough to form a spanning cluster. A typical configuration is shown in Fig. 4(b).

In Figs. 5(a)—5(e) we show the time development of the configurations quenched to $G_1$ in the gel region. For early times [Fig. 5(a)], the small clusters begin to grow. At intermediate times, many of the clusters merge to form larger clusters [Figs. 5(b) and 5(c)]. A spanning cluster forms at $t=20$ [Fig. 5(d)] and in the "long"-time limit ($t=25$) the spanning cluster has absorbed the smaller clusters [Fig. 5(e)]. If the quench is deep into the gel, region, e.g., $G_2$, the clusters merge in beadlike structures (see Fig. 6). At later times the larger clusters absorb the intermediate-sized clusters and continue to grow by filling in the space between clusters until one large spanning cluster forms. Very small clusters, which occasionally break off from the spanning cluster, remain for all times. At early times the peak of the corresponding cluster-mass distribution [see Fig. 3(b)] shifts to larger mass and decreases its height with time. At intermediate times the peak grows but its position remains unchanged. Finally at later times all medium-sized clusters are absorbed by the spanning cluster, and the peak shifts back to smaller sizes. For deeper quenches the system reaches equilibrium more quickly and the identity of the beads forming the clusters becomes more evident.

On the critical surface the dynamical behavior is similar to that observed in the gel phase except that the beadlike structures which form the larger clusters are less discernible and the clusters are more ramified with a fractal dimension which is probably that of ordinary percolation clusters. We find a broader cluster-mass distribution [Fig. 3(c)] in comparison to quenches deep within the gel region where only the very smallest clusters can survive once a spanning cluster is formed. Also near the critical surface there is a greater tendency for the spanning cluster to occasionally dissociate into two large but nonspanning clusters which soon merge again.

Since the dynamical processes do not occur over more than one decade of time (equilibrium is reached for $t=30$—40), it is difficult to determine the existence of scaling laws for the dynamical properties. The dependence of $\ln R$ on $\ln t$ is shown for the largest cluster in Fig. 7 for the gel phase, critical surface, and sol phase. The $t$ dependence of $R$ in the gel phase suggests that the largest cluster initially behaves as $t^x$, with $x$ approximately equal to 0.5. Our interpretation of this early-time behavior is that the initial growth occurs by the addition of newly occupied bonds at a constant rate of increase. That is, $dm(t)/dt \sim \text{const}$ and $m(t) \sim t$. Since the clusters are compact, we obtain $R(t) \sim t^{1/2}$. Note that the mechanism for this time dependence of $R(t)$ is not the same as the curvature-driven growth found in spinodal decomposition.[2] For later times the time dependence of the largest cluster appears to cross over to exponential growth. Evidence for this crossover is that as the size of the lattice is increased, the rate of change of $R$ increases much faster than is expected for simple power-law behavior. We interpret the exponential growth as due to the merging of clusters of comparable size since if clusters of average mass $m$ merge, the rate of change of the mass of the largest clusters is proportional to $m$, i.e., $dm(t)/dt \sim m(t)$. This growth mechanism leads to $m(t) \sim \exp(ct)$, and hence $R(t)$ also grows exponentially with time.

On the critical surface the early-time behavior is consistent with $x \approx 0.5$; for later times $x \approx 0.75$. In the sol phase the effective exponent is found to be approximately 0.5, consistent with the dominant mechanism being single-cluster growth rather than merging.

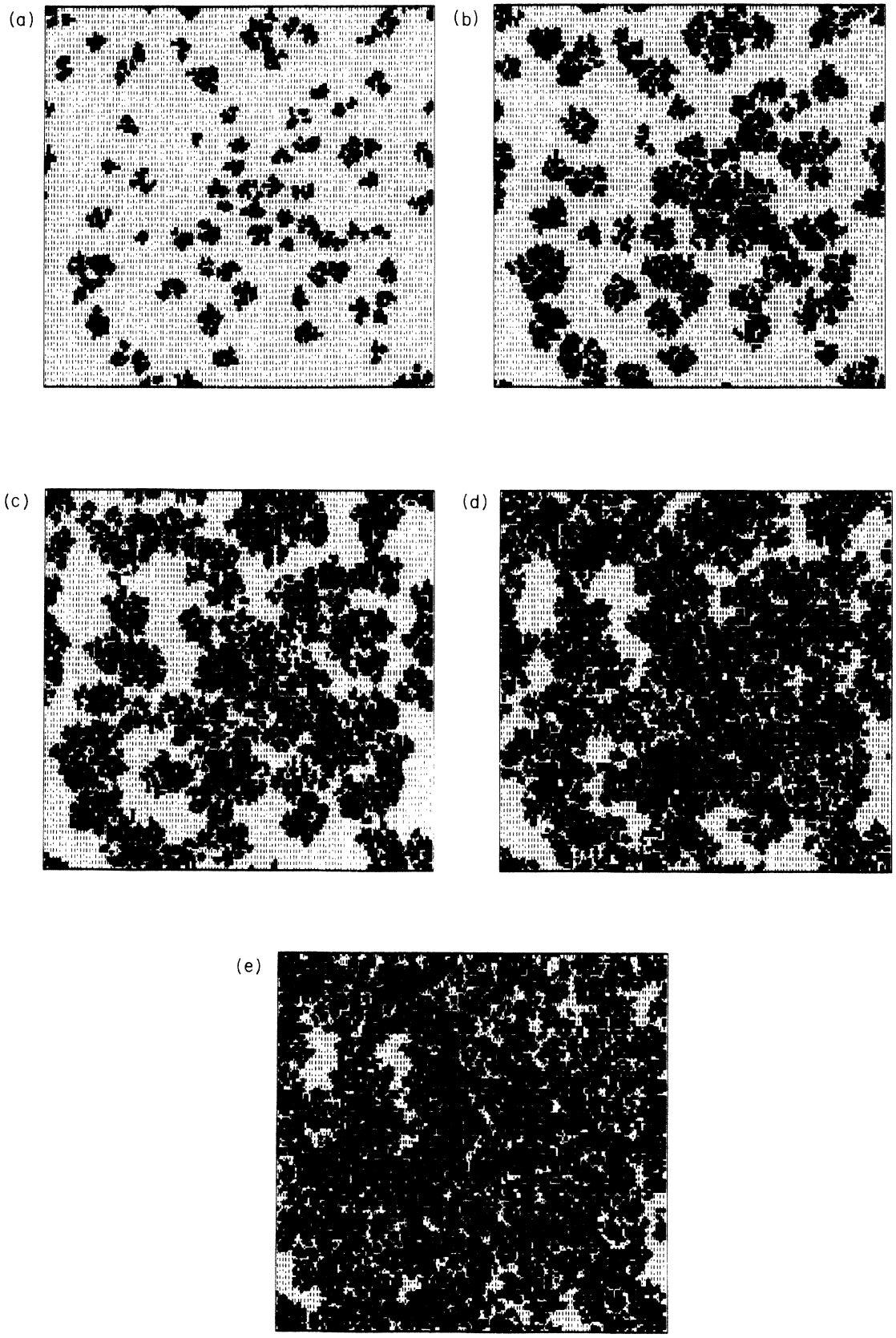


FIG. 5. Time evolution of configurations for the system quenched into gel at $G_1$, where $p_s = 0.8$ and $p_b = 0.8$. (a)—(e) correspond to $t = 5$, 10, 15, 20, and 25 respectively.

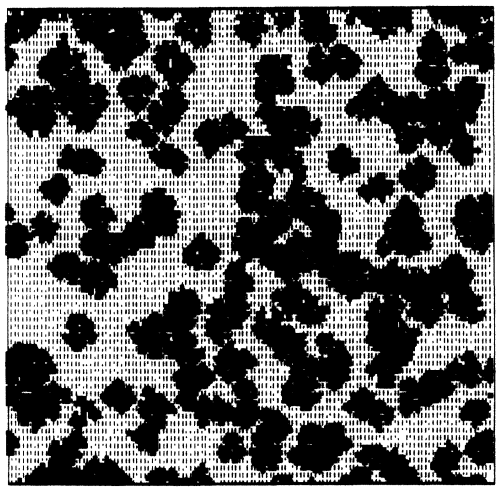

FIG. 6. "Snapshot" of "beadlike" structures at $t=5$ for quench to $G_2$, where $p_s=0.9$ and $p_b=0.8$.

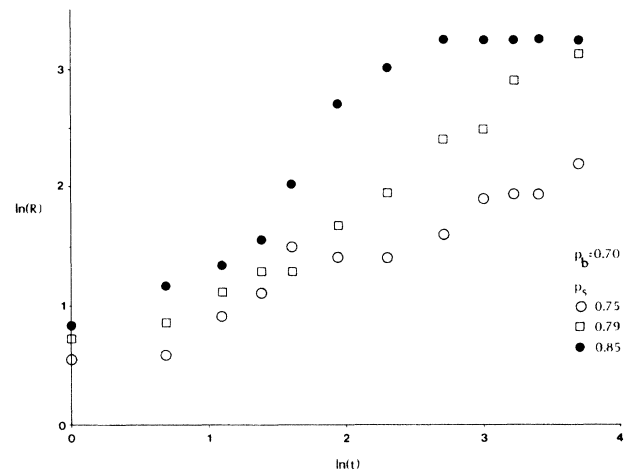


FIG. 7. Log-log plot of the radius of gyration $R$ for the largest cluster versus the number of time steps $t$.

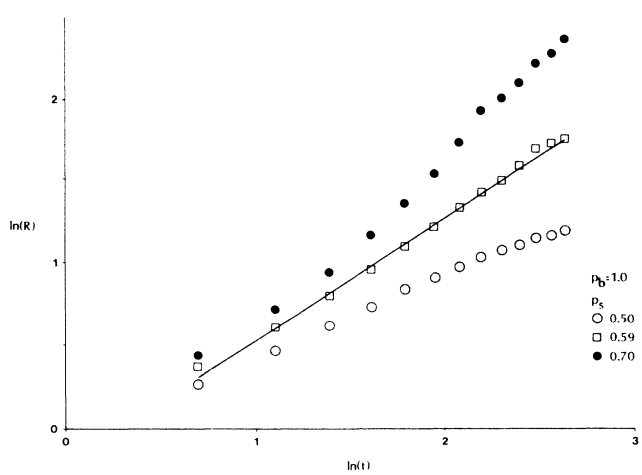


FIG. 8. Plot of $\ln R$ averaged over all clusters versus time in the absence of cluster dissociation.

### C. Absence of cluster dissociation

In the above simulations the sites were tested for occupancy after every time step and excess bonds were culled. We consider here the case for which after the initial quench to probability $p_s$, the sites are not tested again for reoccupancy. For this case the bonds are not culled and the bonds are tested only once for occupancy. Since the clusters cannot dissociate, the behavior of our site-bond dynamics model is easy to understand, and in the long-time limit the static properties of the model are identical to ordinary site-bond percolation. Eventually all occupied sites become part of a cluster or are permanently forbidden from bonding with a cluster. The Monte Carlo results for the critical surface are consistent with Ref. 15 (see Fig. 2). As before, the initial site and bond probabilities are sufficiently low so as to yield only a few small clusters. A quench to a point in the sol region close to the critical surface leads to individual cluster growth for early times and eventual equilibrium in which the clusters have grown to a finite size. For early times after a quench to the gel region, the growth of clusters is by single monomer aggregation. As the mean diameter of the clusters approaches their mean separation, the clusters begin to merge. After a short time one cluster absorbs most of the remaining clusters and spans the lattice. A plot of $\ln R$ versus $\ln t$, where $R$ is the mean radius of gyration of all the clusters, is given in Fig. 8. A quench to the critical surface yields an effective power-law exponent for $R(t)$ of 0.7. For a quench to the sol phase, $R(t)$ increases for early times and then ceases to grow for later times. For a quench to the gel phase, the behavior of $R(t)$ appears to be qualitatively similar to the behavior of $R(t)$ for the largest cluster in the case for which cluster dissociation can occur (see Fig. 7). The reason for this similarity is that for early times the clusters are comparable in size and grow independently; for later times one cluster dominates the average. The slope of $R(t)$ for later times would be greater if the largest cluster only was considered.

As another special case, we consider the Monte Carlo simulation of single-cluster growth as a function of the final site and bond probabilities. For these simulations the lattice chosen was $800\times800$. Our results are consistent with those of ordinary site-bond percolation. We find on the critical surface, $R$ scales at $t^x$ with $x\approx0.80-0.85$. This approximate value of $x$ is consistent with the assumption for this case that the number of time steps scales with the shell number[16] of the cluster. In the gel phase $R(t)$ behaves linearly with $t$ for long times.

## IV. DISCUSSION

We have performed Monte Carlo simulations of a dynamic percolation model quenched into its gel region where the system is unstable to the growth of clusters. This quench from the sol into the gel region corresponds to a critical quench (i.e., in the language of binary alloys, the number of $A$ atoms is equal to the number of $B$ atoms). We find that after the quench the evolution of the system is similar qualitatively to the evolution of a thermal system when quenched into the spinodal or unstable region. In the present model the order parameter (the

probability that a site belongs to the spanning cluster) is not conserved. However, since the instability to the growth of clusters is not due to a long-wavelength fluctuation, we expect that the early-time behavior observed in the present model will be relevant to both the conserved and nonconserved cases which differ only in their long wavelength and long-time behavior.

The evolution of the cluster mass distribution for our model is similar to that obtained by Penrose *et al.*[17] in their computer simulations of binary alloys and Binder and co-workers in their study of spinodal decomposition.[6,18] Although the above work is mostly near the spinodal (in contrast to a critical quench), it is known that there is a gradual change in behavior from the metastable to the unstable region. Penrose *et al.*[17] describe the evolution of clusters as occurring in three stages. In the first stage monomers evolve toward a broader cluster distribution of larger clusters. During the second stage the large clusters continue to grow, but the distribution of small clusters remains roughly constant. In the third stage the number of large clusters decreases, but their size increases; the simulation was not run for a time sufficiently long to produce a spanning cluster. These three stages are similar to the behavior we have obtained for quenches into the gel phase for our model except that we observe a spanning cluster and at later times the number of small clusters again increases. We interpret this latter behavior as due to small clusters breaking off from the surface of the spanning cluster.

Since our model corresponds to a critical quench, it is interesting to compare the configurations we obtain with the "percolation-like" clusters found[19,13] in critical quenches of the Ising model. These structures gradually coarsen and later essentially freeze and grow very slowly. Our configurations at the inception of the spanning cluster qualitatively resemble those generated by simulations of the Ising model. However, comparisons between the qualitative results of our model and the Ising model must be made carefully since, as was discussed in Sec. I, the definition of clusters according to a connected network of nearest-neighbor spins is not adequate. A naive definition of clusters based on nearest-neighbor connectivity that might be used to describe the geometry of the early-time behavior for the Ising-model quench does not have the characteristics found in our model. For example, a quench from very high temperatures, where the spins are uncorrelated, cannot correspond to a large connected group of spins which represent the cooperative behavior of the system. Hence if the clusters were defined appropriately, we expect that the short-time behavior would actually correspond to a few isolated clusters. As the system evolves and the spins begin to be more correlated, the correctly defined clusters would grow and eventually merge as observed in our model. For still later times the cluster picture will either not be useful or the definition of the clusters will change with time.

From the above considerations we believe that the initial growth of clusters which we observe occurs at times before the usual measurements in simulations of Ising-model quenches begin. In addition, after this work was completed, we learned the preliminary results[20] for the Ising model with spin-flip dynamics exhibits two time regimes for times less than one Monte Carlo step per spin. Our data are consistent with theirs. Our results for quenches into the gel region for the growth of the largest cluster show initial power-law behavior and then exponential growth due to merging of clusters of comparable size. The Cahn-Hilliard theory[2,11] predicts exponential growth for the structure function $S(k,t)$, although simulations of the Ising model have not observed this behavior except for long-range interactions.[21] The exponential behavior is predicted[22] to occur only for times on the order of the log of the range of the potential, which for nearest-neighbor interactions is near zero. One of the important aspects of our simulation is that we have been able to extend the early-time regime to observe exponential behavior.

One important issue in the study of spinodal decomposition is the long-time scaling behavior [2,13] of $S(k,t)$. Our calculations are not immediately relevant to the long-time behavior of $S(k,t)$. From our simulations it appears that soon after the spanning cluster is formed, any further growth is very slow.

The evolution of the clusters in the gel region depends on the depth of the quench. Near the sol-gel threshold, there are many neighboring clusters which merge to form a spanning cluster together with a broad distribution of clusters of smaller sizes. On the other hand, for deep quenches, medium-sized beadlike structures appear to grow first which then string together to form the gel. We conjecture that this phenomena might be important in the structure of actual gels. Although several models of the kinetics of gelation have been proposed,[1,23] the kinetics of the unstable sol phase has only recently been considered.[24,25] In this region the ability of the percolation clusters to disassociate and reform drastically changes their behavior in the region far from the gel threshold.

The results found here for our dynamical percolation model raise several problems which we plan to investigate in future work. Our immediate goal is to obtain a definition of clusters for spinodal decomposition in thermal systems which yields a description of cluster growth for early times which is consistent with our qualitative picture. Problems of interest in the context of dynamical percolation-type models include the establishment of universality classes, the determination of their detailed properties including the nature of the cluster interface, the formulation of geometrical models with conservation law analogs, and the theoretical formulation of a cluster-growth model which is isomorphic to spinodal decomposition or thermal instabilities. We are also investigating the formulation of a time-dependent percolation theory based on a generalization of the relation of static percolation to the continuum Potts model.

## ACKNOWLEDGMENTS

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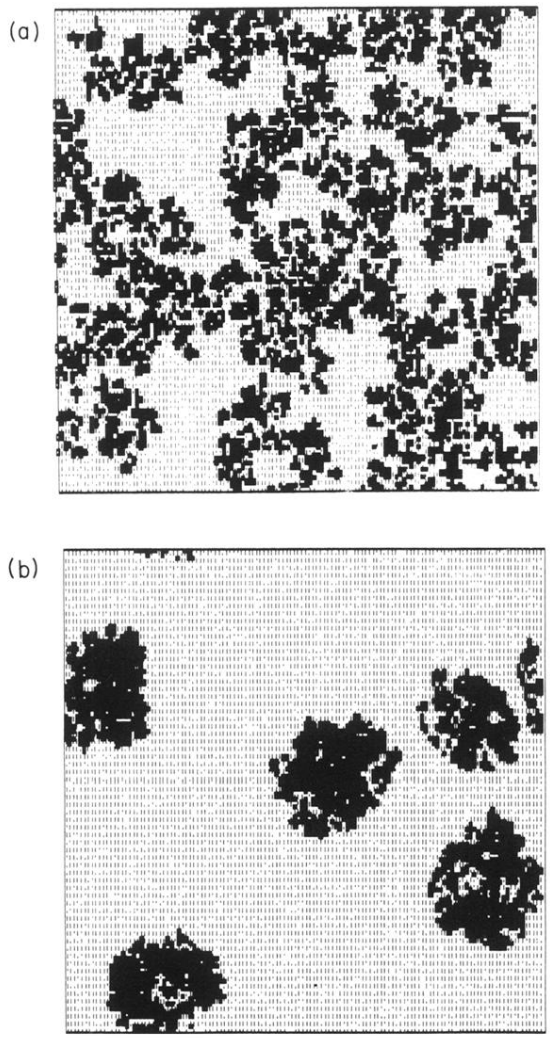


FIG. 4. Snapshots of configurations after 30 time steps for the system quenched into sol phase. (a) corresponds to $S_1$, where $p_s = 0.7$ and $p_b = 0.75$, and (b) corresponds to $S_3$, where $p_s = 0.9$ and $p_b = 0.5$.

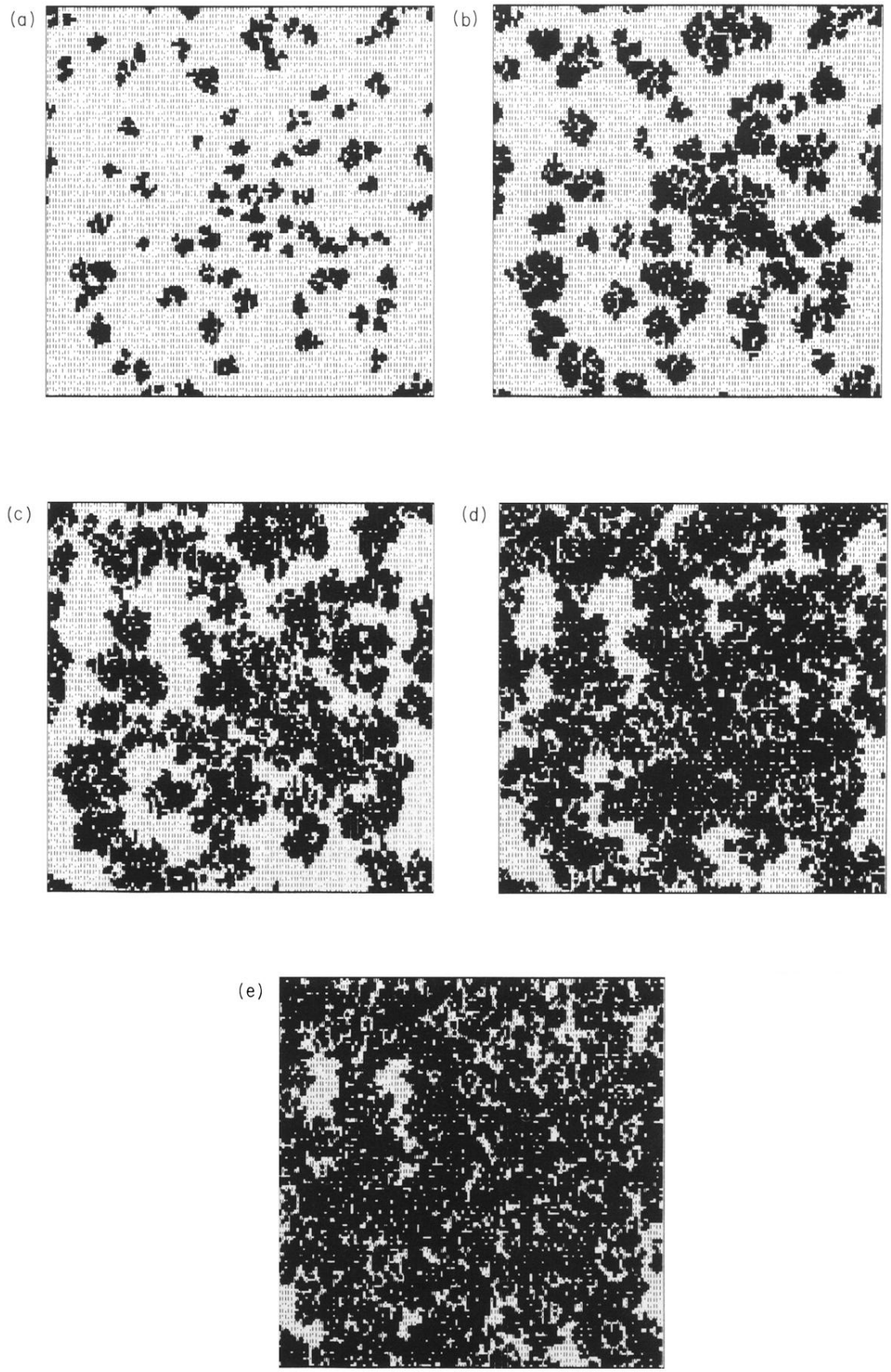


FIG. 5. Time evolution of configurations for the system quenched into gel at $G_1$, where $p_s = 0.8$ and $p_b = 0.8$. (a)–(e) correspond to $t = 5$, 10, 15, 20, and 25 respectively.

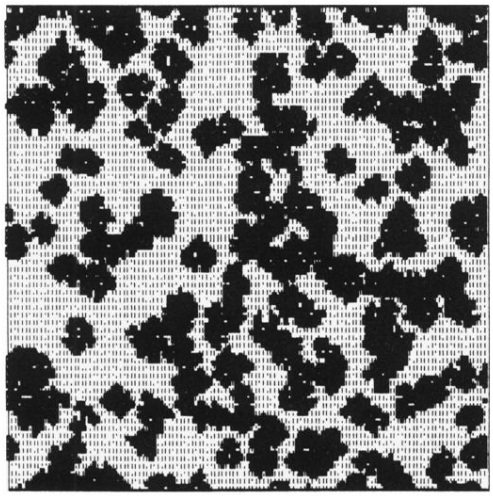

FIG. 6. "Snapshot" of "beadlike" structures at $t=5$ for quench to $G_2$, where $p_s=0.9$ and $p_b=0.8$.