Simulations of a stochastic model for cluster growth on a square lattice

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An irreversible stochastic model for the growth of clusters on a square lattice is formulated and studied by Monte Carlo simulation. The growth rate has a nonlinear nonlocal dependence on the density of occupied sites, qualitatively similar to that of crystallization governed by surface tension and diffusion. The density and Hausdorff dimension of the clusters are estimated. These results are compared with data for percolation clusters and with recent results for diffusion-limited growth.

Growth of crystals from an undercooled melt or a supersaturated solution,[1] coagulation of smoke particles,[2] and growth of cell colonies in a nutrient medium[3] are all examples of growth processes governed by surface tension and diffusion. The surface tension favors compact clusters with a minimal surface to volume ratio. The growth rate at any point is limited by the effective rate of diffusive transport in its neighborhood. Since surface sites deep inside the cluster are connected with the surrounding medium only through relatively narrow passages, the growth rate at such screened sites is reduced, relative to sites on protruding parts of the surface. This screening effect acts on a length scale l_D, which is proportional to the diffusion constant in the medium. The diffusion process thus favors treelike, or dendritic growth. The theory of dendritic growth has been studied extensively by Langer and Müller-Krumbhaar.[1] Recently Witten and Sander[2] have presented results of computer simulations of a cluster growth process where diffusion on a lattice is treated exactly, but where surface tension is ignored. The resulting clusters are extremely branched and have a very low density of occupied sites.

Here we present results of Monte Carlo simulations of the irreversible growth of clusters on a two-dimensional square lattice. The growth process is modeled by an irreversible Markov process defined by an occupation rate $v(\underline{x})$ at the site $\underline{x}$. The rate has a nonlinear nonlocal dependence on the density of occupied sites, defined by

$$v(\underline{x})=A[1-s(\underline{x})]n(\underline{x})\mathscr{D}(\underline{x};l_D)\ . \tag{1}$$

Here $s(\underline{x})=1$ if $\underline{x}$ is occupied, and zero if empty. The term $[1-s(\underline{x})]$ ensures that no site is occupied more than once. $n(\underline{x})$ is the number of occupied nearest-neighbor sites surrounding $\underline{x}$. In fact, $-n(\underline{x})$ is proportional to the discrete analog of surface curvature and corresponds to the surface tension term in the model of Langer and Müller-Krumbhaar. This term also restricts growth to surface sites. The term $\mathscr{D}(\underline{x};l_D)$ corresponds to an effective diffusion constant, expressing the above-mentioned screening effect at $\underline{x}$ due to occupied sites $\underline{x}'$ inside a neighborhood of radius l_D about $\underline{x}$. The particular form used in this work is obtained as a product over square shells surrounding $\underline{x}$ of effective diffusion constants, weighted by an exponential decay on the length scale l_D, namely,

$$\mathscr{D}(\underline{x};l_D) = D\prod_n \exp[-\langle s(\underline{x}')\exp(-|\underline{x}-\underline{x}'|/l_D)\rangle_n]\ , \tag{2}$$

where $\langle\ \rangle_n$ denotes the average over sites $\underline{x}'$ in the nth such shell.[4] If the density of occupied sites is zero everywhere, this expression equals the diffusion constant D in the medium. It is virtually independent of the density at $|\underline{x}'-\underline{x}|>l_D$, but is strongly reduced if the density is large for $|\underline{x}'-\underline{x}|<l_D$. In fact, the detailed analytic form should be rather unimportant as long as this qualitative feature is retained. An alternative definition is

$$\mathscr{D}(\underline{x};l_D) = D[1-l_D^{-1}\sum_n \langle s(\underline{x}')\exp(-|\underline{x}-\underline{x}'|/l_D)\rangle_n], \tag{2'}$$

which is the result of averaging the local diffusion

constant $[1-s(\underline{x})]D$ over space, using

$$l_D^{-1}\exp(-|\underline{x}-\underline{x}'|/l_D)$$

as a weighting factor. This factor is the gradient of the diffusion field in front of a plane cluster surface at $\underline{x}$.[1] A is a normalization constant which does not affect the resulting cluster geometry. It has been chosen so as to keep the computation speed relatively high.

The competition between the stabilizing effect of $n(\underline{x})$, which acts on the length scale chosen as the lattice constant, and the destabilizing effect of $\mathscr{D}(\underline{x};l_D)$, which acts on the length scale of l_D, is qualitatively analogous to that which occurs in cluster growth controlled by surface tension and diffusion. It is therefore to be expected that just as in the latter case, smooth cluster surfaces should be unstable towards dendritic growth for l_D above some critical value.

The Monte Carlo simulations were performed for values of l_D ranging from 0.06 to 10.0 lattice constants. The maximum number of occupied sites ranges from $N=5000$ in the former case, to $N=1500$ in the latter. The speed of the simulation algorithm, which decreases sharply as l_D increases, sets a practical upper limit for N. Each cluster was grown from a seed of one occupied site.

The qualitative changes in cluster structure as l_D is increased are shown in Figs. 1–4. For $l_D<1.0$ the cluster is compact and spherical (Fig. 1). For

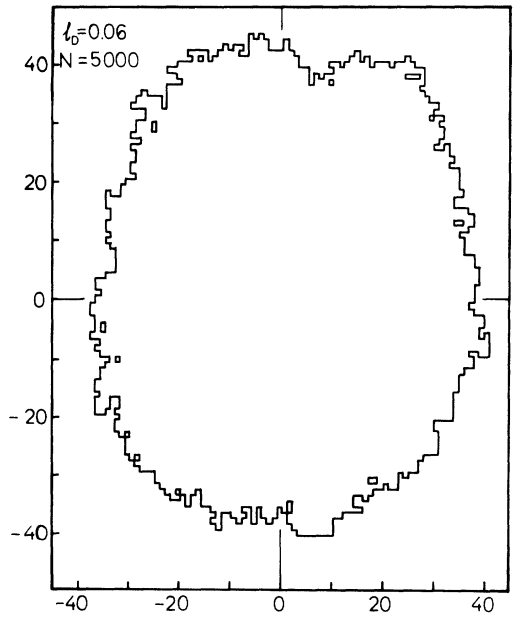

FIG. 1. Cluster of $N=5000$ occupied sites for $l_D=0.06$. Compact, circular shape.

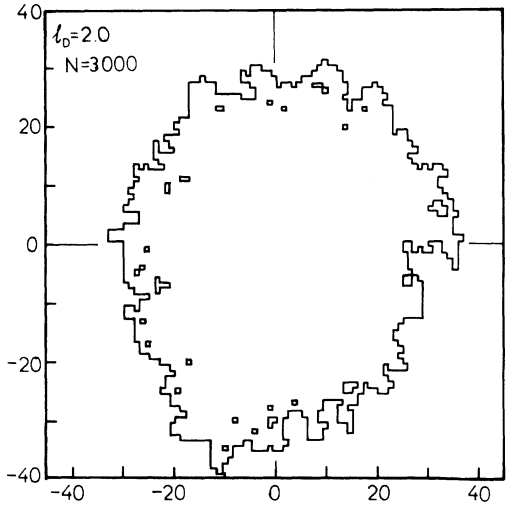

FIG. 2. $l_D=2.0$, $N=3000$. Compact, circular shape with rough surface.

$l_D=2.0$ the surface is rougher (Fig. 2). For $l_D=4.0$ there is a porous structure with a rough outer surface and finite vacancy density in the bulk (Fig. 3). For $l_D=10.0$ the vacancies have coalesced, and the resulting structure is treelike (Fig. 4). There is some suggestion that the system passes through a roughening transition at $l_D\approx 2$, above which there is a finite density of vacancies in the bulk. There may also be a percolationlike transition of the vacancies at $l_D\approx 8$, leading to treelike clusters. Because of finite-size effects it is, however, imposible to decide whether or not these are indeed sharp transitions.

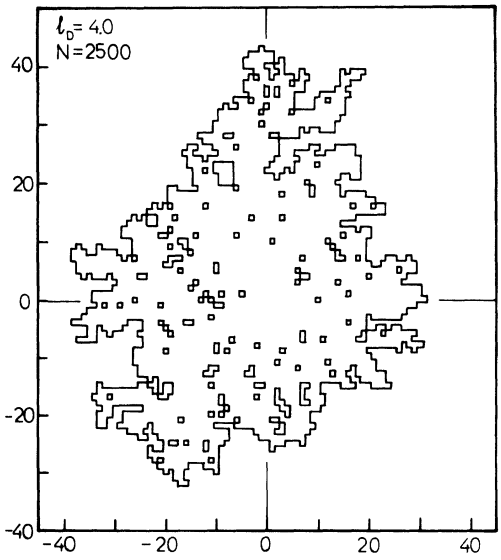

FIG. 3. $l_D=4.0$, $N=2500$. Porous cluster with rough surface and nonzero vacancy density.

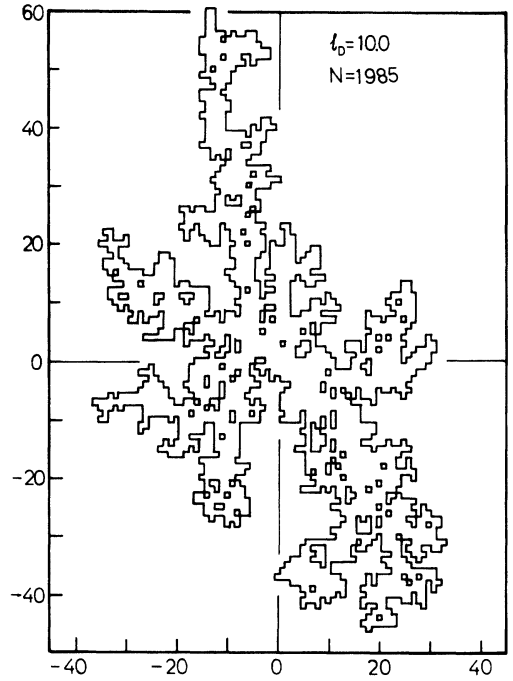

FIG. 4. $l_D=10.0$, $N=1985$. Treelike cluster with mostly surface sites.

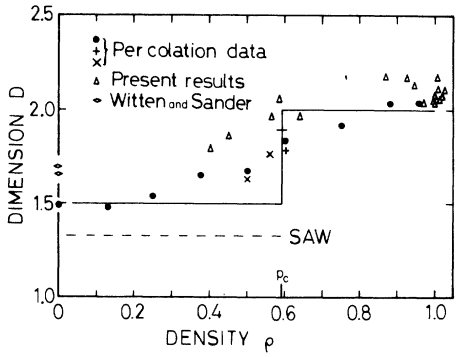

FIG. 5. Hausdorff dimension D vs density ρ (Δ). Also shown are exact and simulation results for site percolation, from Fig. 14 of Ref. 5 ($\cdot, +, \times$). $p_c=0.593$ is the critical density for site percolation on a square lattice. Values $D=2$ for $\rho>p_c$ and $D=2/(1+\frac{1}{18})$ for $\rho=p_c$ are believed to be exact, while $D=1.5$ for $\rho<p_c$ is from simulations. Witten and Sander's results for growing clusters, $D=1.701\pm0.02$ from analysis of radius of gyration, and $D=1.657\pm0.02$ from analysis of correlation functions, are shown at $\rho=0$ ($\Diamond$). For the self-avoiding random walk $D=\frac{4}{3}$ (broken line).

For quantitative analysis we have performed linear regression estimates of the density ρ of occupied sites inside the cluster, based on the quadratic relation

$$N=4\alpha\langle\Delta_N\rangle+\frac{\pi}{4}\rho\langle\Delta_N\rangle^2 \tag{3}$$

between the cluster size N and the root-mean-square diameter Δ_N. The angle brackets denote averages at fixed N over four independent realizations of the growth process. To reduce finite-size effects, only clusters of $\Delta_N>40$ have been included in the fits. ρ decreases monotonically from unity for $l_D<1$, and passes through 0.5 around $l_D\approx 8$. We have also estimated the Hausdorff dimension D of the clusters[2,5] from the asymptotic relation

$$\langle\Delta_N\rangle\sim N^{1/D}. \tag{4}$$

Finite-size effects due to lower-order terms make the estimated values somewhat high. In Fig. 5, D versus ρ is shown, together with exact and simulation results for percolation clusters and the self-avoiding random walk, taken from Stauffer.[6] The values of D obtained by Witten and Sander[2] for $d=2$ are shown at $\rho=0$. For large ρ, corresponding to small values of l_D, our estimated values of D lie well above the maximum possible value 2. This is probably due to finite-size effects. Bur for ρ below the critical density for site percolation on a square lattice $p_c=0.593$, corresponding to $l_D\gtrsim 8$, D decreases well below 2. The values are consistently higher than those for percolation clusters, but seem to be in reasonable agreement with those obtained by Witten and Sander for clusters growing by a true diffusion mechanism in the limit of negligible surface tension.

Although the present study only treats the diffusion process approximately, it also includes surface tension. Thus we have been able to model the interesting effects on surface stability of the competition between surface tension and diffusion. We have observed qualitative features which suggest transitions between different geometrical cluster structures as the diffusion length is varied, but finite-size effects prevent us from drawing firm conclusions about their nature. Further work on improved models, with special attention to reducing the finite-size effects would be useful.

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