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BRIEF REPORTS

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Lattice model of solidification with mobile impurities in the liquid phase

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A solidification front advancing into a three-dimensional (3D) medium containing mobile impurities is implemented as a model for a semilate stage dynamical process at a first-order phase transition under the isothermal undercooling condition. The approach generalizes the dynamical epidemic model to 3D. The presence of mobile particles shifts the usual probability of the percolation transition for 3D systems from 0.65 corresponding to static hindrances, to a value numerically estimated as 0.8. Excluded volume effects caused by the impurity particles lead to an aggregation process self-organizing the particles trapped behind the front in the solid phase. The kinetics of the process early stages is studied numerically and a power law governing the front width evolution is suggested. [S0163-1829(98)06525-4]

I. INTRODUCTION

A model of growth starting from a surface and the statistical physics of the microstructure and surface roughness is studied here through numerical simulation. The problem is, as seen on a general basis, that of a solidification front advancing into a three-dimensional (3D) medium containing mobile impurities, i.e., a rather semilate stage process at a first-order phase transition between three phases and under an isothermal undercooling condition.[1] The work applies to many realistic cases since it considers a (chemically nonreacting) front progressing into a 3D medium containing mobile impurities in interaction with the front. The microstructure is going to be mainly described at the mesoscopic (''particle'') scale rather than at the chemical level. Thick-film growth, polymer growth in impure solutions, fluid invasion in porous media, heterogeneous crowd traffic, heterogeneous granular flow in random medium, etc., are all cases of pertinent interest.

The process model is based on the kinetic Eden model.[2] The space in which the cluster develops is, however, partially filled with mobile ''impurities.'' During the growth, the surface of the front expands, and the impurities are rejected or trapped by the solidifying matrix following a probability rule. A dynamical repulsion effect on the front with the mobile ''particles'' is included as a realistic element. The situation has been investigated numerically. Possible local anisotropy conditions and chemical effects have not been included.

Based on the recent extension of the Eden model[2] to a planar dynamical epidemic model,[3,4] a simple, spatial, dynamical, epidemic model equivalent to the growth of a solid (S) phase advancing into another one (a liquid phase, L) containing mobile, so-called impurities or ''particles'' (P) can be implemented in 3D. A percolationlike transition is expected, and is hereby shown to occur as a function of impurity fraction (Sec. II), i.e., a transition separating regimes of indefinitely growing and nongrowing S clusters. The critical value $x_c = 0.8$ is much above the threshold value $x_c = 0.65$ of the static epidemic model on a cubic lattice[5,6] and also above the dynamical and static values of the corresponding process on a square lattice.[4] The difference is explained as a result of self-organization of the impurities and by the larger possibilities of that process to take place in the space (Sec. III). Moreover the description of the front indicates the possible development of facets, and competition between facets of different orientations with surface roughening, i.e., an interesting microstructure complexity. Excluded volume effects caused by the impurity particles lead to an aggregation process self-organizing the particles trapped behind the front in the solid phase. Some mean-field argument is given in order to qualify the process as in Ref. 7.

The geometrical and time-dependent properties of the front and the aggregates are discussed in Sec. IV.

II. MODEL

Consider a cubic symmetry semi-infinite 3D lattice slab with a square base of size $W \times W$. The z axis is perpendicular to the base. Let periodic boundary conditions hold on the four sides of this square ''substrate.'' Each site of the 3D lattice is supposed to be occupied by a liquid L unit, a solid phase S unit, or by an impurity particle P. At the start of the process, a fixed fraction x of P impurities is supposed to be randomly spatially distributed in the liquid phase region. The solidification process begins with a creation of a substrate, i.e., turning all sites on the first layer from a liquid L to a solid phase S to initiate the cluster growth. At each subsequent simulation step, all liquid L sites in contact with the ''cluster'' are selected. One of them is randomly chosen and is turned into the phase S. Such a growth scheme is equivalent to the classical Eden one.[2] Then the dynamics of impurities is governed by the same rules as stated in Ref. 3. Briefly speaking, a dynamical interaction is assumed to occur between the cluster front and the impurities such that if an impurity is in nearest-neighbor contact with the newly added solid unit, the impurity executes a single lattice step random walk towards one of the nearest liquid site, if any is available, thereby displacing the liquid in order to reduce the contact with the solid front. If the move cannot be made, the position of the impurity remains unchanged and it is trapped in the cluster, e.g., there are two ways in which an impurity particle can be trapped during the processes: (i) it can be trapped directly by the front because the former one cannot reduce its number of nearest-neighbor solid units, or (ii) the displacement of an impurity can be forbidden by the presence of other impurities on neighboring sites, leading to indirect trapping. The selection, front, and impurity motion are randomly chosen many times. If the particle does not move after that process has been repeated 50 times, it is assumed to be trapped. Indeed, the cluster growth can sometimes stop if there is no site in the liquid in contact with the cluster because a cluster of particles lays along the front.

Each of the results, in particular profiles, stems from an average over 500 simulations. For larger sizes W of the system the simulation time becomes very long and the results do not improve much in the process.

The present model is in line with other well-known droplet growth processes[1] and reduces to an Eden model giving compact S clusters,[2] when there are no impurities, and to the simple epidemic model when static (quenched) impurities are considered.[5,6] Other related works can be found in Refs. 8–14. All pertain to isothermal undercooling processes.

III. MORPHOLOGY OF SOLID AND TRAPPED PHASES

Numerical results of the advancing front morphology in the direction perpendicular to the substrate are first discussed. The evolution of the density profiles of the S and P phases at a distance h from the substrate is studied. A typical pattern of the trapped phase by the solid phase cluster and of the advancing front morphology for impurity fraction $x = 0.3$ ($x < x_c$) is shown in Fig. 1 with the linear size of the square substrate taken to be $W = 16$ for illustration. One should note that the distribution of P impurities in the S matrix is quite inhomogeneous behind the front [Fig. 1(a)]. Figure 1(c) illustrates the random distribution of the free impurities which are not yet captured by the solidification process.

The evolution of density profiles for $x = 0.3$ of the impurities and of the solid phase can next be studied in the three mutually perpendicular symmetry directions x, y, z, for the purpose of discussion. The density profile along a given direction and at a certain point is given the contribution of P/S units in the plane perpendicular to the direction at that point. Since it is a spatial model we looked first at the P and S profiles in the x and y directions. They are found to be almost constant and are thus not shown. However, a specific behavior of the impurity density profile $\rho_{P_\perp}(h)$ along the advancing growth in the z direction at distance h from the substrate is observed near the position of the front [Fig. 2(a)].

For all studied density profiles for impurity fractions $0 < x < x_c$: (i) in the first stages of the growth, $\rho_{P_\perp}(h)$ is very

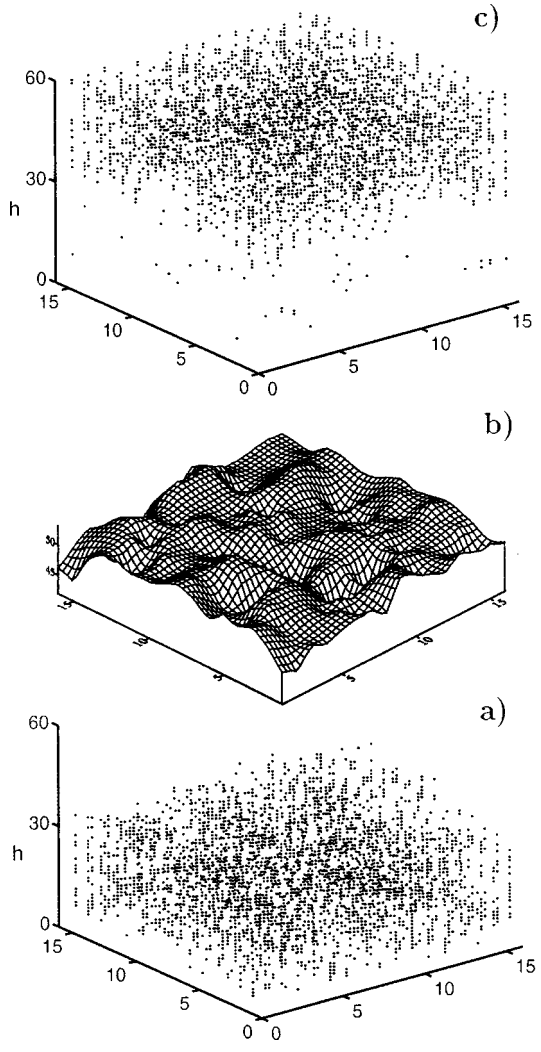

FIG. 1. Typical pattern: (a) of trapped impurities; (b) of the advancing front morphology and; (c) of free randomly distributed impurities for impurity fraction $x = 0.3$ and for a $W = 16$ size of the square substrate.

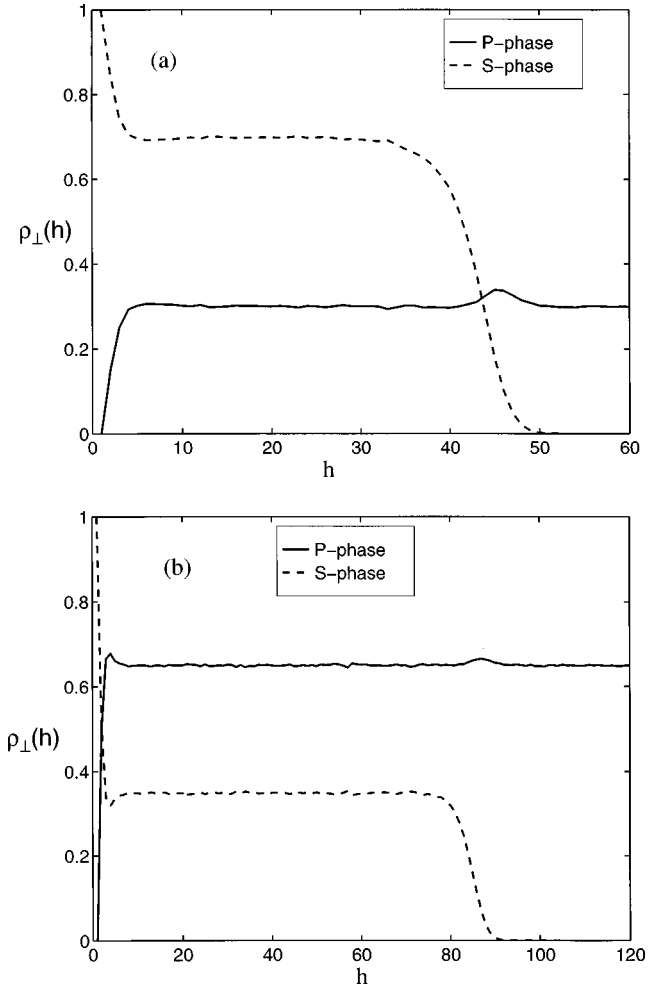

FIG. 2. Profiles $\rho_{\perp}(h)$ for both P and S phases are illustrated as a function of the distance h from the substrate for impurity concentration values: (a) $x=0.3$ and (b) $x=0.65$. The profiles result from an average over 500 simulations.

low near the substrate, and (ii) a bump is clearly seen in the $\rho_{P_{\perp}}(h)$ distribution around the front position, as in the case of a 2D lattice.[3]

The depletion in the $\rho_{P_{\perp}}(h)$ distribution near the substrate can be understood as resulting from the relatively smaller number of constraining hindrances in the early stages of the interface growth. The bump observed in the $\rho_{P_{\perp}}(h)$ profile is an excess of P impurities moving with the front as long as $0<x<x_c$. As the front advances, the bump width Δh increases and the bump height $\Delta\rho$ decreases.

The numerical analysis of the bump height was done through the measurement of the maximum height of the $\rho_{P_{\perp}}(h)$ profile. By fitting the first eight points it was found that the bump height is scaled as a power law with an exponent $\beta=0.317\pm0.038$. The bump height is found to be x dependent as in the 2D case. Numerical results for the bump height obtained for the case of a $16\times16\times30$ cubic lattice dimensions are shown in Fig. 3. The maximum of the parabolalike dependence is observed for impurity fraction around $x=0.3$. Then the curve drops to zero at $x_c=0.8$.

For impurity fractions near the critical value a ripple occurs behind the front just after the main initial depletion

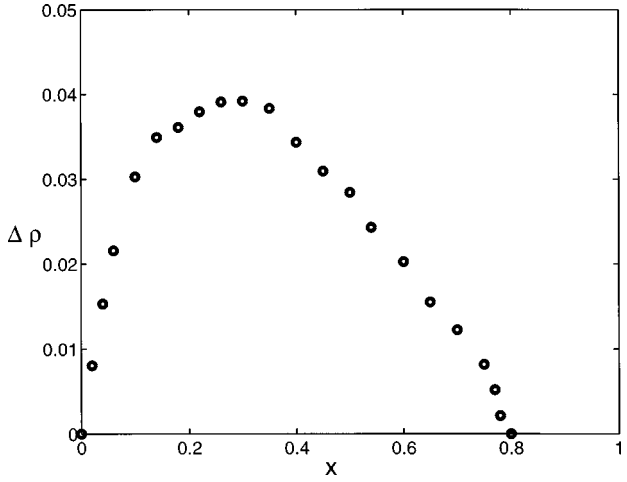

FIG. 3. The x dependence of the bump height $\Delta\rho$.

layer near the substrate [see Fig. 2(b)] as it was observed in the plane and the same schematical presentation holds. This observation can be interpreted and understood as resulting from the approach to fractality of the front, and the resulting trapped impurities.

IV. THE TIME DEPENDENCE OF THE WIDTH OF THE FRONT

The width σ of the front, or equivalently the correlation length ξ in the z direction along the front was numerically studied at the initial stages of the cluster growth process. The correlation length in the z direction is usually defined by[3]

$$\xi=\sqrt{\frac{1}{N}\sum_{i=1}^{N} z_i^2-\left(\frac{1}{N}\sum_{i=1}^{N} z_i\right)^2}, \tag{1}$$

where N is the number of the solid phase units which form a front at distance h (see Fig. 1) from the substrate and z_i is the z coordinate of the position of each of these units. The results for the time evolution of the width show a power-law dependence $\xi\sim h^{\alpha}$ characterized by $\alpha=0.526\pm0.01=1/D$ ($D=91/48$) since it is a 2D structure.[15] After that the correlation length enters a saturation regime (Fig. 4).

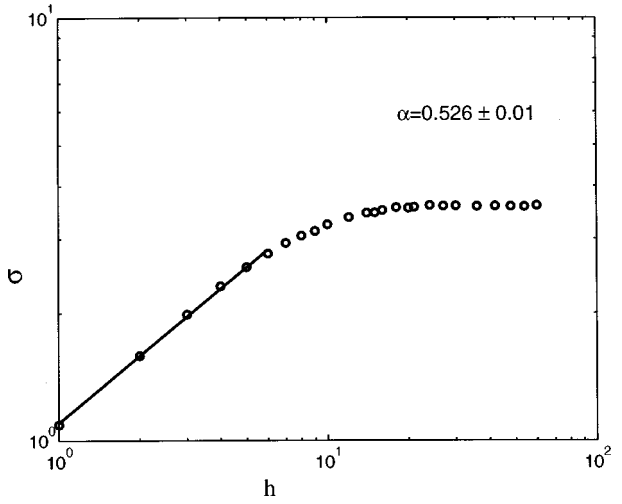

FIG. 4. The evolution of the solidification front width (σ or ξ).

V. CONCLUSION

I have studied the growth of a cluster at a first-order phase transition between three phases along the lines of the dynamical epidemic model for a 2D interface interacting with mobile impurities in a semi-infinite 3D medium, thus as a model for a semilate stage dynamical process at a first-order phase transition under isothermal undercooling condition. During the growth, the repulsion between the front and the impurities leads to an aggregation process along the solidifying matrix, and to a self-organization of aggregates near and after the interface for $0<x<x_c$. The percolationlike transition for a 3D dynamical system has been numerically investigated and found to be much above the one for the corresponding planar structure. That is understood to result from the richer complexity of the rough surface/interface in the way of self-organization.

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