Inner Patterns and Front Propagation of a Dynamic Random Impurity Model

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We investigate a simple model, the so-called dynamic epidemic model, for the evolution of an advancing interface through a medium containing mobile impurities. A short range repulsion between the front and the impurities leads to an aggregation process along the front, and to the trapping of aggregates. The amplitude of this self-organization for *collective* impurity trapping is analyzed in two dimensions. A relationship exists between individual motion and the aggregation of particles. The results indicate that self-organizing features are different behind and on the front. [S0031-9007(96)00607-2]

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Pattern formation is a common phenomenon in nature. Various physical processes leading to spatial and/or temporal self-organization have been investigated, e.g., coalescence, aggregation, convection, and fragmentation, in quite different random systems ranging from polymers to traffic flow. However, less attention has been paid to the trapping of impurities pushed by an advancing interface. This case is of high interest, for example, in crystal growth where there is often decoration of a crystal by impurities. While the pushing or trapping of a *single* impurity by a smooth interface has been experimentally and theoretically investigated [1], there are few studies about the more complex phenomenon of *collective* motion or trapping of many "impurities" near an interface.

We discuss a simple, dynamic, epidemic model [2] equivalent to the growth of a solid phase B advancing into another one (liquid phase, L) containing *mobile* (so-called A) impurities. The latter ones are either pushed by the interface due to some repulsion or trapped by the advancing front. This process leads to the aggregation of impurities along the front and to the trapping of aggregates in the solid phase. A percolationlike transition between growing and nongrowing B clusters is found to take place at an impurity fraction $x_c \approx 0.56$ much above the threshold value $x_c^{(e)} \approx 0.41$ of the static epidemic model on a square lattice [3]. This difference is explained as a result of the *self-organization* of the impurities. The geometrical properties of the front and the aggregates are discussed here. Moreover, features due to such a front motion process as observed in physical and chemical systems can be explained. Applications to various types of interfaces (unmixing fluids, solid-liquid crystallization, noncoagulating liquid droplets or vesicles, etc.) are obvious. Many other spreading phenomena (epidemia, dielectric breakdown, gelation, crowd motion, etc.) can also be of interest for the present study.

The model is constrained to be the simplest possible on a two-dimensional square half-space lattice of width W. Periodic boundary conditions are used. Each site can receive at most an L unit, a solid B unit, or an A impurity. Initially, all sites are of the L phase type except for a fraction x of sites which contain an A impurity, for which the spatial distribution is supposed to be initially random. The growth starts from a seed B line. At each simulation step, all liquid sites in contact with the so-called "cluster" (B) are selected. One of them is randomly chosen and is turned into the phase B following the trivial reaction $L \rightarrow B$. Such a growth rule is equivalent to the classical Eden one [4]. A dynamical interaction is assumed to occur between the front and the impurities such that, if an impurity is in nearest neighbor contact with the newly added solid unit, the impurity makes a random walk towards one of the nearest "liquid" sites, if any are 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 becomes trapped by the front in the cluster. One should note that an impurity can be trapped following two different routes: (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 further "indirect impeachment." The selection, front, and impurity motion process are repeated a large number of times *if possible*. Indeed, the growth can sometimes stop if there is no site in the liquid in contact with the cluster. The growth dynamics is irreversible (i.e., far from equilibrium) and history dependent (i.e., non-Markovian). One should observe that the present model is reduced to well known models for two particular cases: (i) *Without* impurities ($x = 0$), the model reduces simply to an Eden model giving *compact* B clusters, and (ii) the model reduces to the simple epidemic model when *static* impurities are considered [3] and at which percolation transition $x_c^{(e)}$ the B clusters are *fractal* (with a fractal dimension $91/48$) [5]. The present model thus spans a wide open gap in statistical mechanics. A change in the lattice symmetry or in the jump law is expected to give

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the same qualitative but not quantitative results. We have thus constrained the model to the most standard one: nearest neighbor interactions on the square lattice.

Numerical results of the front profile in the direction perpendicular to the substrate are first discussed. The evolution of the densities $\rho_A(h)$ and $\rho_B(h)$ of, respectively, the A and B phases at a distance h from the substrate is shown in Fig. 1 for $x = 0.20$ together with the density profile for the A and B phases. One should first note that the distribution of A impurities in the B matrix is *inhomogeneous* behind the front. The density profile for $\rho_B(h)$ is roughly flat around $1 - x$ and falls abruptly to zero near the front position but with a finite width. The density profile of $\rho_A(h)$ informs us that the concentration of trapped impurities in the B matrix as well as in the L phases is roughly equal to x. However, for $x \approx x_c = 0.56 \pm 0.01$, a sharp modification in behavior exists: The density profile $\rho_B(h)$ behind the front is not flat and is found to follow a power law with an exponent $2 - D_f$. Moreover, the distribution of trapped A particles is also found to be fractal at x_c with the same fractal dimension $D_f = 91/48$ [2].

However, some important deviations from the above observations in the density profile have to be mentioned for $0 < x < x_c$: (i) In the first stages of the growth, $\rho_A(h)$ is very low near the substrate, and (ii) a bump is clearly seen in the $\rho_A(h)$ distribution around the front position.

The depletion in the $\rho_A(h)$ distribution near the substrate is understood as resulting from an easy and frequent pushing process in the early stages of the interface growth. The bump observed in the $\rho_A(h)$ profile is an excess of A 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. It is like a damping out solitary wave motion. The height of the bump $\Delta\rho$ is numerically estimated through the measure of the maximum height of the $\rho_A(h)$ profile and is found to scale as a simple power law $\Delta\rho \sim t^{-\beta}$ with an exponent $\beta = 0.32 \pm 0.02$ close to $1/3$. This value seems to be independent of the impurity fraction in the range of interest $0 < x < x_c$ and is independent of the lattice width W. The bump size is x dependent. The curve $\Delta\rho(x)$ is shown (Fig. 2) to present a maximum around $x = 0.15$ and to shrink to zero at $x = x_c$.

In order to study the *displacement* of the impurities with the front, we have measured the distribution $n(\delta)$ of displacements δ of trapped impurities, i.e., the differences between the positions of the impurities before and after trapping. We found the exponential distributions

$$n(\delta^{\parallel}) \approx \exp(-\delta^{\parallel}/\delta_0^{\parallel})/\delta_0^{\parallel}$$

and

$$n(\delta^{\perp}) \approx \exp(-\delta^{\perp}/\delta_0^{\perp})/\delta_0^{\perp} \qquad (1)$$

for all values $0 < x < x_c$ in the parallel ($\parallel$) and perpendicular ($\perp$) directions to the substrate. One should note that the distributions are not the same and a marked *anisotropy* $\delta_0^{\perp} > \delta_0^{\parallel}$ is observed for the impurity motion (Fig. 3) except at $x = x_c$ where both $\delta_0^{\perp}$ and $\delta_0^{\parallel}$ seem to tend to 1. The displacements $\delta_0^{\perp}$ and $\delta_0^{\parallel}$ are found to be a nontrivial function of x (see Fig. 3).

The *quantity* of pushed impurities (i.e., the surface of the bump) is roughly the quantity of trapped impurities lacking near the substrate since the density of trapped impurity behind the front is equivalent to the fraction of

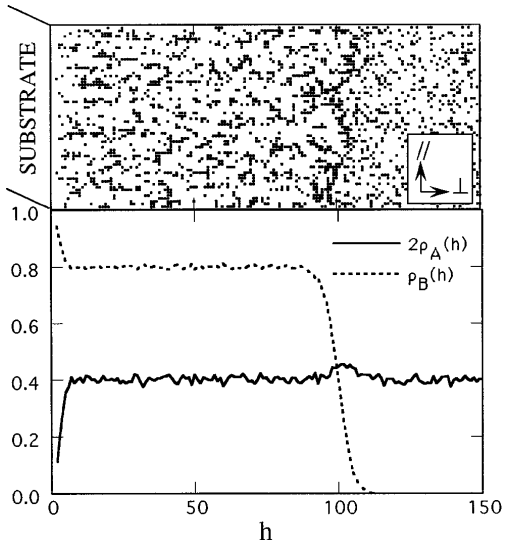

FIG. 1. A typical pattern of width $W = 64$ simulated for $x = 0.20$. Black dots represent A impurities. Profiles for both A and B phases are illustrated as a function of the distance h from the substrate. The profiles are an average over 40 simulations. The scale of $\rho_A(h)$ curve is doubled for clarifying the observation of a bump around the front position.

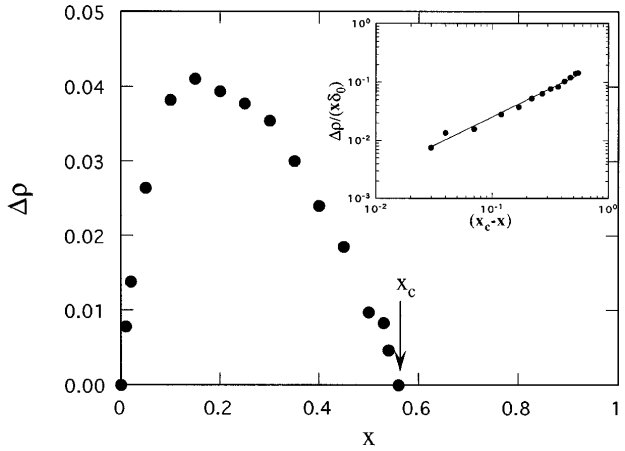

FIG. 2. The x dependence of the bump height $\Delta\rho$. Each dot is an average over 500 simulations. The inset presents a log-log plot of the quantity $\Delta\rho/x\delta_0^{\perp}$ as a function of $x_c - x$. The power law fit is illustrated and gives an exponent $\nu_{\perp} = 1.02 \pm 0.05$.

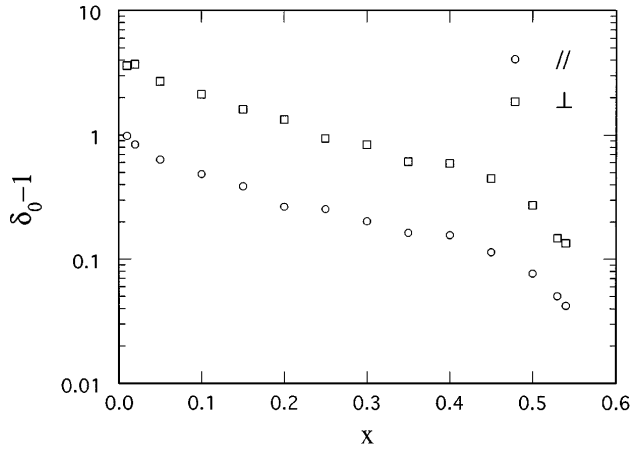

FIG. 3. Semilogarithmic plot of the displacements $\delta_0^{\perp}$ and $\delta_0^{\parallel}$ as a function of the impurity fraction x.

impurities x in the liquid phase. This conservation of the number of pushed impurities is illustrated in Fig. 4(a) through the dashed surfaces which are equivalent. Because the quantity of deficient impurities near the substrate is proportional to $x\delta_0^{\perp}$, one can assume that $x\delta_0^{\perp} \sim \Delta h \Delta\rho$. Moreover, because the bump results from a pushing on the front, the width Δh of the bump could be considered as the width σ of the front. This length is also expected to be the characteristic size of the aggregates which are created and thereafter trapped in the B matrix.

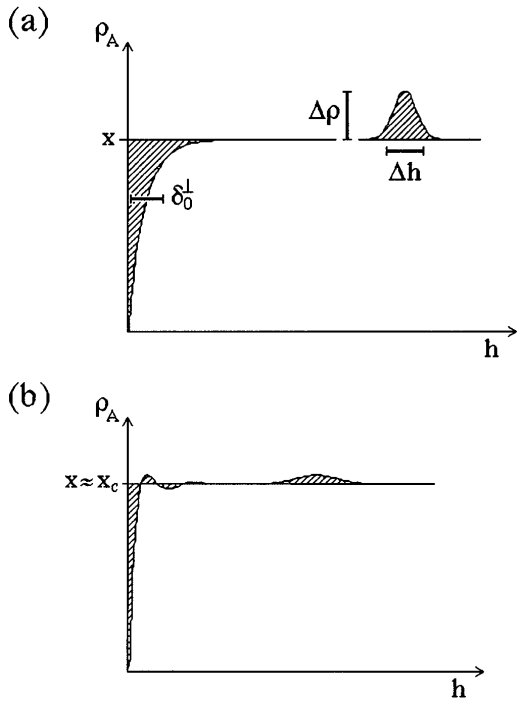

FIG. 4. Schematic representation of $\rho_A(h)$ profile. (a) The various lengths of interest ($\Delta\rho$, Δh, and $\delta_0^{\perp}$) are illustrated. (b) A ripple remains behind the front near the substrate for $x \approx x_c$.

Thus

$$\Delta\rho \sim x\delta_0^{\perp}/\sigma . \tag{2}$$

This relates two independent measures of the aggregation of impurities: the first one following the motion of each individual impurity ($\delta_0^{\perp}$) and the second one for the amplitude of the aggregation phenomenon ($\Delta\rho$).

In percolation [5] and related phenomena [6], the characteristic length of the system (which is σ herein) diverges at the threshold x_c as

$$\sigma \sim (x_c - x)^{-\nu_{\perp}} . \tag{3}$$

This is the same as saying that the size of the aggregates diverges at x_c. This latter physical ansatz then gives for the bump height

$$\Delta\rho \sim x\delta_0^{\perp}(x_c - x)^{\nu_{\perp}} . \tag{4}$$

This inset of Fig. 2 presents the measured quantity $\Delta\rho/x\delta_0^{\perp}$ fitted by a simple power law $(x_c - x)^{\nu_{\perp}}$ where the critical exponent is $\nu_{\perp} = 1.02 \pm 0.05$. This is in contrast with the correlation exponent $4/3$ for the B-cluster length divergence near x_c [2]. This result was checked to be independent of the lattice width W. The results suggest that the correlation lengths of both trapped aggregates and the B front diverge differently at the percolation threshold x_c. This shows that the self-organization is different behind and on the front.

Finally, we have observed for $x \approx x_c$ that a ripple occurs behind the front just after the main initial depletion layer near the substrate as schematically drawn in Fig. 4(b). This observation can be interpreted and understood as resulting from the approach to fractality of the front, and the resulting trapped impurities. This finding should be useful in discussing phenomena observed in chemical reactions in finite systems and in cases like the recently discussed hard sphere distribution near a wall [7].

All these results (aggregation phenomenon on the front, inhomogeneous distributions of impurities, bump evolution, concentration profiles, etc.) are evident of a self-organization process occurring on the growing front for all values, $0 < x < x_c$. The results indicate that self-organizing features are different behind and on the front.

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