Dynamical scaling during interfacial growth following a morphological instability

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The dynamical evolution away from an unstable steady state is studied for the two-sided symmetric model in two dimensions by means of numerically solving the interface equations of motion. Evidence is presented for the appearance of a regime of self-similar growth in which the pattern is characterized by a single length scale $R(t)$. The asymptotic time dependence of such a length is $R(t) \sim t$. The results also show that the local deviation of the interface from planarity, the local normal growth velocity, and the power spectrum of the interface satisfy scaling relations. In addition, by assuming the existence of such a scaling regime, we are able to derive from dimensional and heuristic arguments a power-law growth with exponents in accord with those found in the numerical solution.

I. INTRODUCTION

There has been a great deal of progress and activity in studies of the formation of patterns in various growth processes. Such problems are part of the general question of the behavior of two-phase interfaces under a variety of nonequilibrium conditions. On the theoretical side, they are of interest because of the emergence of complex spatio-temporal patterns. Their study involves dealing with nonlinear and sometimes nonlocal behavior of moving boundaries. On the practical side, questions of crystal growth, hydrodynamic instabilities, and kinetics have important technological consequences. Specific systems that have been under intense study include (i) directional solidification in binary systems,[1,2] (ii) viscous fingering in rectangular[3,4] and circular Hele-Shaw cells including anisotropic cells,[5-11] and (iii) dendritic growth of a solid from a melt.[12] Additional references and background can be found in Refs. 13–15.

Much of the attention on these and related problems has focused on the existence of steady states, their stability, and the circumstances under which the steady states form a continuous or a discrete set. The aim has been and remains, for example, to understand how the tip velocity and curvature of an advancing front during the solidification of a simple fluid are singled out in a steady-state regime.[13,15] It is currently argued in the case of dendritic growth in monocomponent fluids and in the case of finger growth in experiments using Hele-Shaw cells that steady states form a discrete set, out of which a small number (perhaps a single one) is linearly stable. There have been some detailed checked on this scenario, for example in the case of viscous fingering[16] and, quite recently, in directional solidification.[17-19,15,20,21]

Less attention has been paid to date on the nature and dynamics of the growth process itself. Brush and Sekerka[22] have studied the growth of a slightly perturbed circular seed in a supercooled liquid. They were able to follow the growth of the crystal well inside the nonlinear regime and to analyze the evolution of the Fourier spectrum of the interface.

Most other dynamical analyses to date, however, have considered one or another variation of diffusion-limited aggregation, first studied in detail by Witten and Sander.[23] A great deal of information as been obtained about such kinetic processes. These latter studies were, however, not really designed to include the assumption of local thermodynamic equilibrium, which is implicit in the Mullins-Sekerka instability lying at the heart of many interfacial growth instabilities.[24]

To deal somewhat more realistically with local thermodynamics and the concomitant relaxation processes at the interface, Guo and Jasnow[25,26] modeled the case of diffusion of matter by using a lattice gas model and Ising-type interactions among the particles. Monte Carlo simulations were performed to study the dynamical evolution away from an unstable flat interface, when the interface was driven from equilibrium by a steady flux of particles. Conserved (Kawasaki[27]) dynamics was used to model diffusion, and the local Ising interaction allowed one to incorporate relaxation of the interfacial structures so that the additional *ad hoc* introduction of interfacial boundary conditions was not required. Hence the conservation of particles and the so-called Gibbs-Thomson effect were automatically included. The latter follows from the microscopic evaporation-condensation mechanism at the interface.

The simulations of Guo and Jasnow were designed after the symmetric model introduced by Langer and Turski.[28] It is known that such a model in an infinite geometry does not have linearly stable steady states, and thus it allows a detailed study of the transient evolution away from an unstable flat interface. Hence it is important to consider an infinite system (or, in practical simula-

tions, a very large, though finite, system) to avoid the effects of the boundaries. What emerged from these preliminary studies was a scaling regime of self-similar growth.[25] Guo and Jasnow calculated the power spectrum of the interfacial displacement $h(x,t)$ at time t, namely,

$$P(k,t)=|h(k,t)|^2=N^{-2}\left|\sum_{i=1}^{N} e^{ikx_i}h(x_i,t)\right|^2 , \quad (1)$$

where the summation runs over the N sites along the direction parallel to the interface. The power spectrum was shown to have a scaling form

$$P(k,t)=At^{\delta}f(Bkt^{1/z}) . \quad (2)$$

The amplitudes A and B are nonuniversal, but available evidence indicated that the scaling function $f(x)$ is universal, that is, independent of the driving flux and temperature, over the small range of parameters that was accessible. (In the lattice gas model the temperature controls the surface tension.) The implication of Eq. (2) is that the dominant mode in the pattern grows as a power law, i.e.,

$$P(k_{\max}(t),t)\sim t^{\delta} . \quad (3)$$

Furthermore, scaling indicates the existence of coarsening, i.e., a characteristic length scale for the structures which grows in time, $R\sim t^{1/z}$. These forms of scaling appear quite naturally in related studies of spinodal decomposition.[29,30] There, however, one has an ensemble of random interfaces, and the system is not driven externally but, rather, is approaching thermodynamic equilibrium.

The Monte Carlo simulations, while extremely encouraging, have their weaknesses, largely due to the small systems used and the inherent statistical fluctuations. Improved Monte Carlo calculations can be carried out as, for example, in the work by Harris and Grant[31] or by reducing the noise along the lines of work on the Saffman-Taylor problem.[32] Use of the macroscopic interface equations, however, holds out the possibility for more accuracy and the ability to investigate more deeply the nature of the scaling and structures, and to vary, in a more controlled fashion, parameters such as the anisotropy.

In this paper, results of a numerical solution of the macroscopic interface equation for the two-sided symmetric model in two dimensions are presented. The evidence for an asymptotic scaling regime appears to be very good in the nonlinear growth domain. Furthermore, the macroscopic interface equation allows a scaling analysis which is based on the numerical evidence of dynamical scaling. Such an analysis predicts an exponent $z=1$ and $\delta=2z$ for the model studied, in good agreement with the numerical results.

The layout of the remainder of this paper is as follows. In Sec. II the symmetric model and the accompanying interface equation are introduced. Section III is devoted to a presentation of the results of the numerical solution of the interface equation. The final section is reserved for a dimensional analysis of the interface equation and for concluding remarks.

II. SYMMETRIC MODEL

Following Langer and Turski[28] one imagines a system with two ordered equilibrium phases, denoted by α and β, coexisting at a temperature $T<T_c$. The diffusion coefficient and order parameter susceptibility are assumed to be the same in both phases. The system is infinite in extent, and the equilibrium interface is taken to be the plane $z=0$, with the α (respectively β) phase occupying the half space $z>0$ (respectively $z<0$).[33] In each of the two ordered phases, macroscopic order parameter inhomogeneities relax diffusively according to

$$\partial_t\phi_\gamma=D_\gamma\nabla^2\phi_\gamma, \quad \gamma=\alpha,\beta . \quad (4)$$

This equation has to be supplemented with appropriate boundary conditions at the interface. Conservation of order parameter leads to

$$D(-\nabla\phi_\alpha+\nabla\phi_\beta)\cdot\hat{\mathbf{n}}=(\Delta\phi)v_n , \quad (5)$$

where the unit normal $\hat{\mathbf{n}}$ is directed into the α phase, v_n is the local normal velocity, and $\Delta\phi$ is the equilibrium miscibility gap. The appropriate Gibbs-Thomson relation is of the form

$$\phi|_S=\phi^{\mathrm{eq}}+\Gamma\kappa[h] , \quad (6)$$

where the interface shape is given by $z=h(x,t)$, and κ is the mean curvature of the interface, taken as positive if the nearest center of curvature lies in the α phase. The subscript S indicates that the quantity is to be evaluated as the limit approaching the interface from one or the other bulk phases. The coefficient Γ contains thermodynamic information and is of the form $\Gamma=\sigma\chi/\Delta\phi$, where σ is the surface tension and χ is the order parameter susceptibility in the equilibrium phases (equal in both phases in this symmetric system).

We have used in the present study a constant flux boundary condition far from the interface: $\mathbf{j}(z=\pm\infty)=j_0\hat{\mathbf{k}}$, where $\hat{\mathbf{k}}$ is the unit vector in the z direction. This system of equations and boundary conditions admits a steady-state solution in which a flat interface at, say, $z=0$, separates both phases. The order parameter inside either phase is given by

$$\phi^{(0)}=\phi^{\mathrm{eq}}-(j_0/D)z . \quad (7)$$

The values ϕ^{eq} correspond to the bulk equilibrium order parameter; for specificity we take $\phi_\alpha^{\mathrm{eq}}=-\phi_\beta^{\mathrm{eq}}>0$.

We now define a field $u(x,z)$ such that

$$\phi(x,z)-\phi^{\mathrm{eq}}=-(j_0/D)z+(\Delta\phi)u(x,z) . \quad (8)$$

In the symmetric model, u is continuous across the interface. The steady-state solution corresponding to a flat interface at $z=0$ is given by $u=0$. This solution, however, is known to be linearly unstable for perturbations of sufficiently long wavelength.[28,34] In the remainder of this paper we will study the dynamical evolution of the interface from an initially unstable flat interface.

We furthermore introduce the quasistationary approximation in which one neglects retardation effects and replaces the full diffusion equation by Laplace's equation in both phases. This is clearly not a consistent approxima-

tion for all times in an infinite system.[35] However, in any realistic situation, the consistency can be addressed. In any event including retardation effects is certainly beyond the scope of the work reported here.

The system of partial differential equations and boundary conditions described above can be recast into an equation of motion for the interface alone. As has been discussed above, we consider a finite domain of width W, with periodic boundary conditions. We define a "Coulomb" Green function by $\nabla^2 G(\mathbf{r}|\mathbf{r}')=-\delta(\mathbf{r}-\mathbf{r}')$. Since the numerical solution will be obtained in a cell of finite width with periodic boundary conditions, it is convenient to impose periodic boundary conditions on G as well.[17] The explicit form of the Green function reads

$$G(\mathbf{r}|\mathbf{r}')=-\frac{1}{2W}|\Delta z|-\frac{1}{4\pi}\ln[1-2p\cos(k_0\Delta x)+p^2]\ , \tag{9}$$

where $\mathbf{r}=(x,z)$, $\Delta z=z-z'$, $\Delta x=x-x'$, $k_0=2\pi/W$, and $p=\exp(-k_0|\Delta z|)$. With this choice of Green function, the equation for the interface reads

$$\Gamma\kappa(s)+\frac{j_0}{D}h(s)=\int_S G(s,s')\frac{\Delta\phi}{D}v_n(s')ds'\ , \tag{10}$$

where the interface has been parametrized by a contour variable s. s and s' denote two arbitrary points on the interface and the integration extends over the entire interface.

We have described the interface on a discrete set of points and numerically solved for its motion in the following way. At time t the interface configuration and hence the left-hand side of Eq. (10) are known. Equation (10) is then an integral equation for the normal velocities v_n. Once the normal velocities are found the interface position is determined at a new time $t+\delta t$ by forward integration in time.

The equation of motion determines the normal velocities only; it is useful to add a tangential component to keep the points equally spaced in arc length. We determine this tangential component as follows (the discussion specialized to a two-dimensional system). If $S_{\rm tot}$ is the total length of the interface (imagining, for the moment, a finite or periodic system), we require that the relative position of a point on the contour remains unchanged, i.e., $(s/S_{\rm tot})_t=(s'/S_{\rm tot})_{t+\delta t}$, where s' is the image of the interface point s under the prescribed iteration scheme that determines the interface at $t+\delta t$. In practice the interface is known at a fixed number of nodes so that the condition described implies that equally spaced nodes remain equally spaced under the motion of the interface. Under this condition, the discrete interface equation can be rewritten as a system of ordinary differential equations (ODE) for θ, the angle between the local normal and the z direction, and the total arc length $S_{\rm tot}$:

$$\frac{\partial\theta}{\partial t}=v_t\kappa+\frac{\partial v_n}{\partial s}\ , \tag{11}$$

and

$$\frac{dS_{\rm tot}}{dt}=-\int_0^{S_{\rm tot}}\kappa(s)v_n(s)ds\ , \tag{12}$$

where v_t is the tangential velocity determined by $v_t(s)-v_t(0)=sg(S_{\rm tot})-S_{\rm tot}g(s)$ with

$$g(s)=\frac{1}{S_{\rm tot}}\int_0^s v_n(s')[-\kappa(s')]ds'\ . \tag{13}$$

If the interface is represented by a set of N equally spaced nodes in arc length, Eqs. (11) and (12) constitute a system of $N+1$ independent ODE's. There is one additional equation for the motion of a reference point on the interface. We have integrated the equations of motion with an implicit ODE solver such as HSODEN in the Cray BCSLIB library. We have typically used $N=242$ nodes in our calculations and occasionally increased N to $N=362$.

III. RESULTS

One would like to study the evolution of the interface free from the influence of outer boundaries. The approach taken here is that space is infinite, but that in the direction perpendicular to the direction of the imposed flux, the structures are periodic. In order to minimize further the perturbing influence of the imposed periodicity, we have made special choices of the physical parameters, as described below. This choice of parameters allows us to consider several relevant Fourier modes of the interface while keeping the width of the system, W, reasonably small.

We rescale lengths by the capillary length, $d_0=\Gamma/\Delta\phi$, and times by d_0^2/D. Then the only control parameter is $j_0/D\Gamma=(d_0 l)^{-1}$. The dimensionless length l is inversely proportional to the slope of the order parameter in the steady state [Eq. (7)]. It is known from linear stability theory that the flat interface is unstable against infinitesimal perturbations of wave number $k<k_c=(d_0 l)^{-1/2}$, i.e., the geometric mean of the capillary length and the length l is involved in a characteristic way.[28,34] In real systems the difference between the two lengths may be 5 or 6 orders of magnitude. This fact poses a difficult problem for numerical computations. One hopes, however, that the essential features of the growth behavior can be identified with a much narrower range of lengths. We believe that the essential point is that the structures that evolve must be sufficiently larger in scale than the capillary (or microscopic) length. In this paper results are presented for $l=40$ and $W=800$. The range of operation is fairly narrow, as it was in the original Monte Carlo simulations.[25,26] Increasing l slows down the growth thus increasing the required length of the runs, but decreasing l causes too many modes to be linearly unstable thus requiring a finer interface discretization than is practical. Given our choice of parameters, modes $n=1-20$ are linearly unstable, where, as usual, the wave numbers are specified by $k=2\pi n/W$, with n a non-negative integer. The fastest growing modes in the linear regime are $n=11$ and 12.

In our calculations we have used two types of initial conditions. The first one consists of a linear combination in k space of most of the modes that are linearly unstable, including the dominant modes of the linear theory. [The shortest wavelength ($n\geq 17$) unstable modes can cause numerical difficulties but grow slowly. These are re-

moved from the initial conditions.] The complex coefficients in the linear combination are chosen at random according to a Gaussian distribution with zero mean and width equal to one. With this choice the maximum initial amplitude of any mode is about 2% of its wavelength. The results are then averaged over an ensemble of initial conditions. With this type of initial condition we have observed dynamical scaling of the type which will be described below. Nevertheless, the power spectrum of the interface is peaked, even at reasonably long times, around $n=11$ or 12. This is due to the fast growth of these modes during the linear regime. Of course, this part of the power spectrum is not expected to satisfy dynamical scaling, and its large amplitude masks somewhat the behavior of other modes of longer wavelength. As a consequence we have also used a different type of initial condition designed to minimize the effect of the linear regime and thus to make the nonlinear regime more readily accessible to the numerical calculation. This second type of initial condition is identical to the first one except in that initial amplitudes for modes $n \geq 11$ are strongly filtered, for example, with a sharp cutoff. The results for the power spectrum and different measures of a characteristic length of structure have been obtained with the second type of initial condition and include averages over ten independent runs. Available evidence seems to indicate that the long-time behavior of the system is the same regardless of what type of initial condition has been used. This point, however, will have to be addressed more precisely in future work.

In order to test the accuracy of the numerical scheme, we have analyzed the linear growth regime immediately after the instability. The different interface modes grow independently, with rates in very good agreement with those predicted by linear stability analysis.

In Fig. 1 a typical interface profile is shown for different times. To give immediate circumstantial evidence for a scaling regime we consider three independent lengths: (i) the change in the total arc length, $\Delta S = S_{\rm tot}(t) - S_{\rm tot}(t=0)$, (ii) the mixing zone ΔZ defined as the maximum peak-peak height in the pattern, and (iii) $z_2 = \langle z^2 \rangle^{1/2}$. the rms displacement of the interface about the initial position, $z=0$. In Fig. 2 we show the proportionality of these lengths at sufficiently long times, indicating that there is statistically one characteristic length for the spatial pattern. (The proportionality is equally evident in the individual runs, a situation quite different from simulations of spinodal decomposition which require an average before any systematic behavior emerges.) Furthermore, consider one particular run, like the one shown in Fig. 1. In Fig. 3 we show that for times longer than $t=1500$, all curves collapse onto a single curve if the displacement $z(s)$ is scaled by the characteristic length ΔZ and the arc length is scaled by $S_{\rm tot}$. This is very impressive collapse, clearly not evident at earlier times. Furthermore, the same behavior is observed by plotting the normal velocity $v_n(s)$ versus $s/S_{\rm tot}$. (The choice for the scaling of the normal velocity will be discussed below.) Finally, the growth of the total arc length versus time is shown in Fig. 4. We conclude that $\Delta S \sim t$, at sufficiently long times.

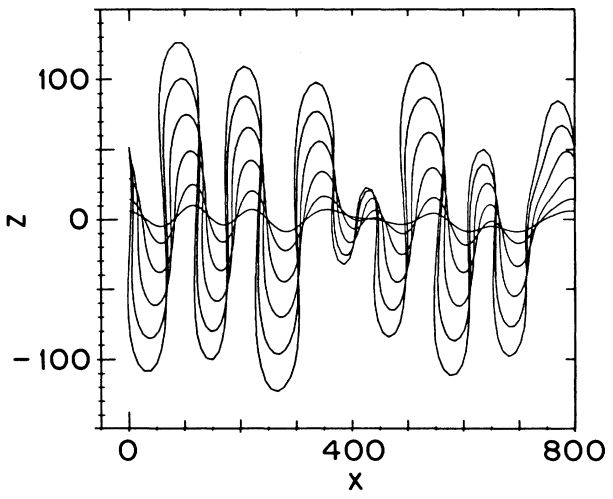

FIG. 1. Typical sequence of interface profiles (in x-z coordinates) following the instability of a planar interface. The width of the system is $W=800$ and the times shown are $t=0$, 500, 1000, 1500, 2000, and 2500, in order of increasing maximum amplitude.

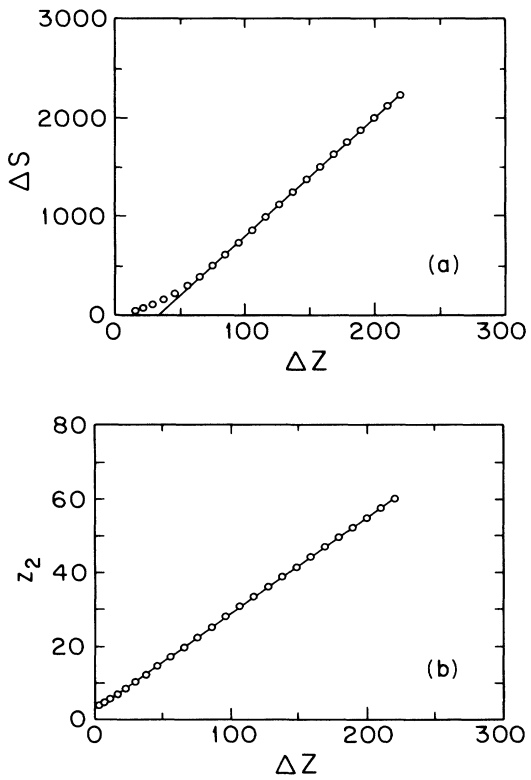

FIG. 2. (a) Change in the total arc length, ΔS, as a function of the maximum peak-to-peak amplitude, ΔZ, averaged over ten runs. After an initial transient, these two independent lengths become proportional to each other, signaling the existence of a scaling regime. (b) Root-mean-square displacement of the interface, z_2, as a function of ΔZ, averaged over ten runs.

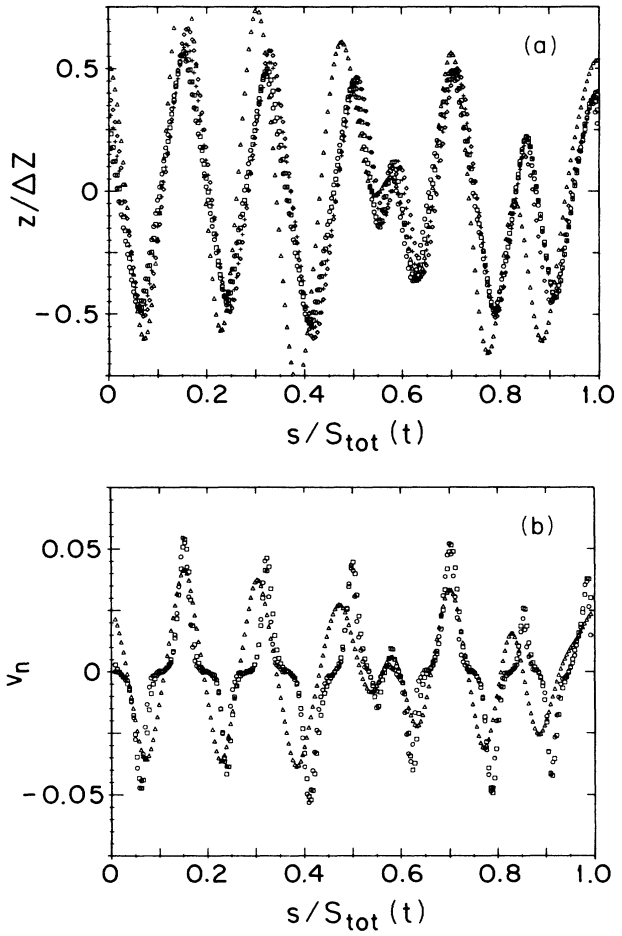

FIG. 3. (a) Rescaled interface profile at several times after the instability: $z/\Delta Z$ vs $s/S_{\rm tot}$. The times shown are △, $t=500$; ◇, $t=1000$; +, $t=1500$; □, $t=2000$; and ○, $t=2500$. At late times, all curves superimpose indicating the existence of a self-similar mode in the dynamical evolution of the interface. (b) Rescaled normal velocities at several times after the instability: v_n vs $s/S_{\rm tot}$. This scaling is consistent with $\beta=0$, as described in the text. The times shown are △, $t=500$; □, $t=2000$; and ○, $t=2500$.

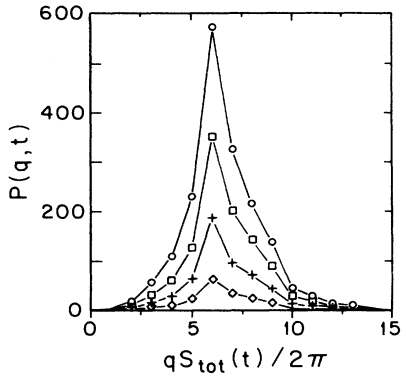

FIG. 5. Power spectrum of the interface averaged over ten independent runs. The initial condition used in each of these runs assigns zero initial amplitude to the modes with $n \geq 10$. The times shown are ◇, $t=1000$; +, $t=1500$; □, $t=2000$; and ○, $t=2500$.

In related studies of, for example, spinodal decomposition, a power-law behavior in a characteristic length scale can be observed before an accurate scaling of the dynamic structure factor has been established. A similar situation has been encountered in the present case. Because of reentrant shapes, we have considered the power spectrum of the displacement $z=h(s)$ as a function of the arc length. The Fourier modes then correspond to cycles in the total arc length, so that

$$P(q,t)=N^{-2}\left|\sum_{i=1}^{N} z(s_i)e^{iqs_i}\right|^2, \tag{14}$$

where the sum is over the equally spaced nodes defining the interfacial contour. The averaged power spectrum is shown in Fig. 5; note the abscissa determines the number of cycles in the total arc length. In Fig. 6 the power spectrum is scaled by the square of the characteristic length ΔZ and the wave number is again scaled by $S_{\rm tot}$. Note that the last times seem to converge to a single curve.

The circumstantial evidence presented indicates that the growth pattern following this form of a Mullins-

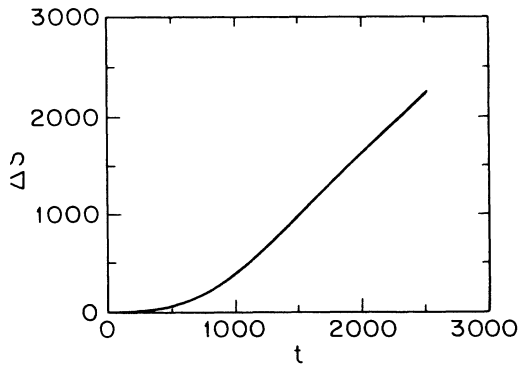

FIG. 4. Time dependence of the change in the total arc length, ΔS, as a function of time t averaged over ten independent runs. Both are proportional at sufficiently long times.

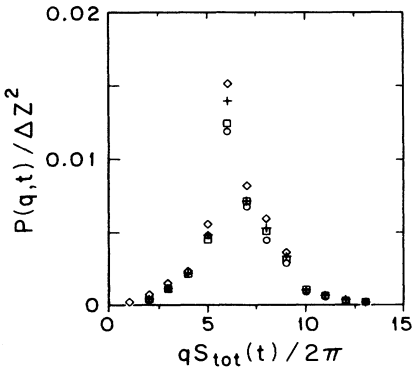

FIG. 6. Power spectrum shown in Fig. 5 scaled with ΔZ. The same times are shown. The curves corresponding to the two latest times are seen to converge.

Sekerka instability selects a self-similar mode in the dynamical evolution. In the next section we discuss the scaling from the point of view of the interface equations of motion.

IV. SCALING OF THE INTERFACE EQUATIONS AND DISCUSSION

We present in this section an analysis of the scaling properties of the interface equation. Our analysis is based on the empirical observation of dynamical self-similarity described in the previous section. We follow the approach given by Mullins and Viñals[36] for the study of coarsening in spinodal decomposition.

Briefly, we assume that after a possible transient, the system is statistically self-similar; i.e., consecutive configurations of the structure are geometrically similar in a statistical sense. As a consequence, any parameter of the structure that is invariant under a uniform length rescaling is also independent of time. Consider the rate of change of the total arc length,

$$\frac{dS_{\rm tot}}{dt} = -\int_0^{S_{\rm tot}} \kappa v_n ds \ . \tag{15}$$

Now suppose that under a uniform length rescaling by λ, the local normal velocity of an element of interface in the scaled system, v_n^λ, is related to the normal velocity of the corresponding element of interface in the original system by $v_n^\lambda = \lambda^\beta v_n$. Then we can write

$$\frac{dS_{\rm tot}}{dt} = \Lambda S_{\rm tot}^\beta \ , \tag{16}$$

where

$$\Lambda = -\frac{1}{S_{\rm tot}^\beta}\int \kappa v_n ds \tag{17}$$

is invariant under length rescaling and, according to our hypothesis, is also independent of time in the self-similar regime. Equation (16) can be integrated to give ($\beta < 1$)

$$S_{\rm tot}^{1-\beta}(t) - S_{\rm tot}^{1-\beta}(t_0) = (1-\beta)\Lambda(t-t_0) \ , \tag{18}$$

where t_0 is some initial time in the self-similar regime.

We proceed now to determine the value of β from the interface equation (10). Imagine that at a given time the interface shape is $z = h(x,t)$. The local normal velocities can be calculated from Eq. (10). Under a uniform rescaling of the linear dimensions of the system by λ, we can write, for the scaled-up system,

$$\Gamma\kappa^\lambda + \frac{j_0}{D}h^\lambda = \frac{\Delta\phi}{D}\int G^\lambda(\lambda s, \lambda s')v_n^\lambda ds^\lambda \ . \tag{19}$$

This equation determines the local normal velocities in the scaled-up system v_n^λ. Since $G(s,s')$ scales as λ^{d-2}, where d is the space dimensionality of the system, $\kappa^\lambda = (1/\lambda)\kappa$ and $h^\lambda = \lambda h$, we obtain

$$\Gamma\frac{1}{\lambda}\kappa + \frac{j_0}{D}\lambda h = \frac{\Delta\phi}{D}\int G(s,s')v_n^\lambda \lambda\, ds \ . \tag{20}$$

For sufficiently large λ (or, equivalently, sufficiently long times), the first term in the left-hand side will be negligible compared to the second. Comparing Eqs. (10) and (20) we find $v_n^\lambda = \lambda^0 v_n$, or $\beta = 0$.

We show in Fig. 3(b) v_n versus $s/S_{\rm tot}(t)$ for different times after the instability. After some transient time, all the curves superimpose indicating that $\beta = 0$, as derived above. We also shown in Fig. 5 ΔS versus time. After an initial transient, ΔS is linear in time, also in agreement with $\beta = 0$. [See Eq. (18).]

Hence the assumption of a self-similar growth regime allows one to extract some details from the equations of motion which are seen to be consistent with the actual simulations. The evidence is good that there is indeed a scaling solution to which the system evolves. There is as yet no theoretical structure within which to understand the existence of scaling. The situation is quite analogous to that of spinodal decomposition, and we believe, because of similarities at both the macroscopic and microscopic levels, that progress in the understanding of coarsening in both bulk and interfacial problems will be related.

There are several additional directions that work on the interfacial scaling domain should take. It will be necessary to examine the one-sided model in which the diffusion coefficients for the two coexisting phases are taken to be widely different. Preliminary indications from Monte Carlo simulations[25,26] suggest different classes of behavior. Second, the entire question of the effect of a force field biasing diffusion in the two phases needs to be considered. Such a field played an important role in the Monte Carlos simulations, and indications are that there are interesting conceptual points involved both at the Ginzburg-Landau level[37] and at the level of macroscopic interface equations. Additionally we note the inherent inconsistency of treating an infinite system in the quasistationary approximation, i.e., replacing the full diffusion equation for the bulk with Laplace's equation. As argued elsewhere[35] the retardation implicit in the diffusion equation may make the treatment of interface equations with a cutoff Green function of interest. These and other directions are currently being explored and will be discussed separately.

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