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Universal features in growth kinetics: Some experimental tests

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Experiments are proposed to test the degree of universality in the long-distance and long-time growth characteristics of ordering unstable systems. Theoretical predictions are also offered that can be scrutinized in these tests.

I. INTRODUCTION

In the area of growth kinetics[1-9] there has been considerable speculation about the existence of universality with respect to the long-distance and long-time growth characteristics. In this article I propose some experimental tests which I believe can help to sort out this question. I also offer some theoretical predictions which can be scrutinized in these tests.

If the ordering in a unstable system is governed by a single scalar order-parameter field $\Psi(\mathbf{R},t)$, the physically relevant observable is the order-parameter correlation function

$$C_\Psi(\mathbf{R},t)=<\Psi(\mathbf{R},t)\Psi(0,t)> , \tag{1.1}$$

where t is the time after a rapid quench from a stable disordered region of the phase diagram to an unstable region of the phase diagram. In particular, the structure factor $C_\Psi(\mathbf{q},t)$, which is just the spatial Fourier transform of $C_\Psi(\mathbf{R},t)$, is measured in scattering (x-ray or neutron) experiments. The first interesting qualitative piece of information which came out of simulations and experiments[10-12] was that $C_\Psi(\mathbf{R},t)$ and $C_\Psi(\mathbf{q},t)$ satisfy the scaling relations

$$C_\Psi(\mathbf{R},t)=F_\Psi(\mathbf{R}/L(t)) , \tag{1.2}$$

$$C_\Psi(\mathbf{q},t)=L^d(t)f_\Psi(\mathbf{q}L(t)) , \tag{1.3}$$

where d is the spatial dimensionality of the system. The scaling function f is found experimentally to be approximately independent of t and the final temperature T_F for a variety of systems[13-39] including liquid mixtures, glassy systems, and binary alloys. The physics of this problem is that there is a single length $L(t)$ which dominates at long time and distances. It is natural to associate $L(t)$ with the average domain size which increases steadily as the system equilibrates.

The key questions are the following.

(1) What are the "growth laws" $L(t)$ and scaling function $F(x)$ for various particular systems?

(2) Are there groups of systems which share the same growth law and scaling functions? Is there a degree of universality?

A simple and rather universal picture has emerged for the growth law $L(t)$ as confirmed by numerous simulations[40-65] of kinetic Ising and Langevin models.[66-72] For pure systems with a nonconserved order parameter (NCOP), the domain growth is curvature driven and the growth law is given by the Lifshitz-Cahn-Allen[73,74] result $L\sim t^{1/2}$. For pure systems with a conserved order parameter (COP) but no hydrodynamic couplings,[75] the growth law is given by the classical Lifshitz-Slyozov[76] result $L\sim t^{1/3}$. If the system is impure[77-86] then one can have logarithmic growth.

In the absence of general analytic methods, much less has been done on quantitatively analyzing the scaling function $F(x)$. Gunton and his group[87] have carried out simulations on a variety of systems, compared the structure factors, and found qualitative similarity. Ohta, Jasnow, and Kawasaki[88] (OJK) carried out a very clever approximate calculation for $F(x)$ for a NCOP system but it has not, to my knowledge, been used[89-93] to determine $F(x)$ for a COP system and it appears difficult to generalize since it contained an uncontrolled approximation involving the statistical distribution of interfaces.

The question remains: What does the scaling function $F(x)$ depend upon? If it is universal we should determine it once and for all and move on to a new set of problems. However, if there are various parametric dependences, then this might be used to extract some interesting information about various ordering systems.

II. DIRECT VISUAL MEASUREMENT OF $F(x)$

Most of the experimental work on growth-kinetics problems have involved scattering experiments which directly measure the structure factor $C(\mathbf{q},t)$. Given the development of modern direct visualization techniques, it seems worthwhile to try and motivate some direct measurements of $C(\mathbf{R},t)$. In this paper, I discuss the type of information one can access in such measurements and how they can be definitive with regards to the question of universality.

The measurements are simple in principle. If one can "see" the contrast between ordering phases, then one can make a "picture" of $\Psi(\mathbf{R},t)$ which assigns an amplitude to a discrete set of N points $\mathbf{R}$ in space. These variables can then be correlated to obtain, at a particular time,

$$C(\mathbf{R},t)=\frac{1}{N}\sum_{\mathbf{R}_1}\Psi(\mathbf{R}+\mathbf{R}_1,t)\Psi(\mathbf{R}_1,t) .$$

The main technical problem is to obtain threedimensional snapshots of $\Psi(\mathbf{R},t)$ or to properly analyze two-dimensional slices. For discussion of "direct observation" in the case of a phase-separating fluid see Ref. 38.

Let us suppose, and I leave it to the experimentalist to devise the most clever technique for such an observation, that one can determine $C(\mathbf{R},t)$ as a function of $\mathbf{R}$ and t in this manner. I want to focus on the particular spatial regime where this data can be represented as a power series in R:

$$C(\mathbf{R},t)=C_0+C_1R+C_2R^2+C_3R^3+\cdots \ . \tag{2.1}$$

The main point I want to make here is that there is conclusive information contained in the time-dependent coefficients C_0, C_1, C_2, and C_3 concerning the growth-kinetic class of a system.

It should be clear that the expansion (2.1) is valid only over a certain restricted set of length scales. Let us assume that we are working at a time after a quench t which is sufficiently large that the typical domain size $L(t)$ is very large compared to the interfacial width ξ. The expansion (2.1) will then hold for distances R small compared to $L(t)$. From an experimental point of view, this is convenient since the range of interest will grow as time evolves and $L(t)$ increases. Let us, for now, assume that we restrict the analysis to the case where R is large compared to the interfacial width ξ. What, then, does one expect for the coefficients C_n? Let me begin with general comments and then be more specific with predictions from a recently developed theory.

III. GENERAL BEHAVIOR OF THE C_n

If, as a quenched system evolves, one develops a set of sharp interfaces,[94–96] then general arguments indicate that the linear coefficient in (2.1) will be nonzero. This is a nonanalytic contribution which leads to Porod's law[94] for the Fourier transform of (2.1)

$$C_\Psi(\mathbf{q},t)\sim q^{-(1+d)} \ . \tag{3.1}$$

If there is scaling then C_1 must be inversely proportional to $L(t)$. On generalizing Porod's law, Tomita[95] has speculated that the coefficient C_2 (and all higher-order even-numbered coefficients) vanishes in this regime. If scaling holds then C_n is proportional to L^{-n}. It has been speculated[96] that these results hold independent of the type of dynamics driving the system.

In the scaling regime[97] the scaling function can be defined by

$$C(\mathbf{R},t)=C_0F(x) \ , \tag{3.2}$$

where $\mathbf{x}=\mathbf{R}/L$. Then, for $x \ll 1$, (2.1) can be rewritten as

$$F(x)=1-\alpha x(1+\beta_2x+\beta_3x^2+\cdots) \ . \tag{3.3}$$

If the "Tomita[96] sum rule" holds, then $\beta_2=0$.

IV. THEORETICAL PREDICTIONS

Let me turn next to the results of a recent theory[98–100] which gives detailed predictions for the scaling function $F(x)$ and the coefficients β_i defined by (3.3). The theoretical model analyzed in this work is the time-dependent Ginzburg-Landau (TDGL) equation

$$\frac{\partial\Psi(\mathbf{R},t)}{\partial t}=-\frac{\Gamma}{2}(-\nabla_R^2)^p\frac{\delta F[\Psi]}{\delta\Psi(\mathbf{R},t)} \ , \tag{4.1}$$

where, on the right-hand side of (4.1), F is an effective Ginzburg-Landau-Wilson Hamiltonian or free-energy functional and Γ sets the time scale. For definiteness, assume F is of the "squared-gradient" form

$$F=\int d^dR\left[\frac{c}{2}(\nabla_R\Psi)^2+V(\Psi)\right] \ , \tag{4.2}$$

where $V(\Psi)$ is the potential, d is the spatial dimension, and $c>0$. In a growth-kinetics context, $V(\Psi)$ is chosen to be a degenerate double-well potential. A simple realization of V is the Ψ^4 potential $V=\frac{1}{4}(1-\Psi^2)^2$. For $p=0$ in (4.1), $(-\nabla_R^2)^0=1$ and this is a form of model A used in dynamic critical phenomena.[101] In this case, the order parameter is not conserved (NCOP) and the model corresponds to, for example, the kinetics of an order-disorder transition in a binary alloy or of an Ising antiferromagnet. For $p=1$ the order parameter is conserved (COP)

$$\frac{d}{dt}\int d^dR\,\Psi(\mathbf{R},t)=0 \ , \tag{4.3}$$

and the kinetics correspond to model B in critical phenomena[101] or the Cahn-Hilliard[102] model in the metallurgical literature. This equation describes spinodal decomposition (phase separation) in binary alloys and Ising ferromagnets. Equation (4.1) is to be supplemented by random initial conditions typically taken to be uncorrelated from site to site and uniformly distributed such that $\langle\Psi(\mathbf{R},t_0)\rangle=0$ and

$$\langle\Psi(\mathbf{R},t_0)\Psi(\mathbf{R}',t_0)\rangle=\delta_{\mathbf{R},\mathbf{R}'}\epsilon_I \ . \tag{4.4}$$

I am assuming in this development that the quench at time $t_0=0$ is to zero temperature where the usual noise term on the right-hand side of (4.1) can be set to zero.

Let me discuss first the results for the NCOP case[99] since it is simpler. Consider the case of quenches to very low temperatures where thermal fluctuations of the order parameter can be ignored. The first important result is that one can express the growth law in the form

$$L(t)=\frac{\xi}{\sqrt{2}}[1-S(t)/\Psi_0^2]^{-1} \ , \tag{4.5}$$

where $S(t)=\langle\Psi^2(\mathbf{R},t)\rangle$ is the order-parameter autocorrelation function, Ψ_0 is the ordered equilibrium value of the field Ψ, and ξ is a measure of the interfacial width

$$\xi=\frac{1}{\Psi_0^2}\int_{-\infty}^{+\infty}[\Psi_0^2-\sigma^2(x)]dx \ . \tag{4.6}$$

In (4.6), $\sigma(x)$ is the exact interfacial profile and Ψ_0 and $\sigma(x)$ can be determined from equilibrium considerations. As the system orders $S(t)\rightarrow\Psi_0^2$, and $L(t)$ increases.

Let us consider the long-time regime where $L\gg\xi$. The theory gives that the correlation function can be written in a scaling form

$$C_\Psi(\mathbf{R},t)=\Psi_0^2 F(\mathbf{R}/L) \ . \tag{4.7}$$

When $x=R/L$ is small, the scaling function can be written in form (3.3) and the theory gives explicit expressions for α and the β_i:

$$\alpha=\left[\frac{2}{\pi(d-1)}\right]^{1/2} , \tag{4.8}$$

$$\beta_2=0 , \tag{4.9}$$

$$\beta_3=-\frac{\mu+\pi/6}{2(2d+1)} , \tag{4.10}$$

where

$$\mu=\begin{cases}1.104 , & d=2 , \\ 0.592 , & d=3 , \\ 0.414 , & d=4 .\end{cases} \tag{4.11}$$

The theory therefore does lead to Porod's law, it does give $\beta_2=0$, and it also gives explicit expressions for the coefficients.

It is nontrivial to obtain even the linear term in (3.3) from a first-principles theory—indeed, with the exception of OJK (Ref. 88) for the NCOP case, no theory up to the present work has been able to reproduce this result. The reason is simply that any mean-field[103–118] theory is incapable of producing such a nonanalytic result. Such theories all give a form

$$C_\Psi(\mathbf{R},t)=\Psi_0^2[1-D(t)R^2+\cdots] \ . \tag{4.12}$$

It is also true that *no* first-principles theory, including OJK, has been able to quantitatively reproduce the expected $t^{1/3}$ growth law for the COP case. The recent work of Bray[119] producing the $t^{1/3}$ growth law contains assumptions which have been questioned by Goldenfeld and Oono.[120] Bray's method cannot, in any event, be used to extract the scaling function. It is worth pointing out that the results stated above do not depend on the form of the potential, the type of underlying lattice, or the nature of the initial conditions [ϵ_I in (4.4)].

Let me turn next to the more challenging[100] COP case. There turns out to be an important physical difference between the NCOP case and the COP case which leads to a subtle theoretical point. In the NCOP case the eventual motion of the interfaces is completely decoupled from the ordered regions in the problem. In the COP case one expects that domain growth is facilitated by the diffusion of material of opposite orientation across ordered domains. Protuburances on an interface serve as a source for fluctuations which break off and propagate across an ordered region. Thus, unlike the NCOP case, one can have fluctuations inside domains which are generated and persist for very long times. This coupling between the interfacial motion and bulk diffusion is necessary to obtain the Lifshitz-Slyozov[76] $t^{1/3}$ growth law.

The determination of the scaling function F is similar to, but more involved than, the NCOP case for what turns out to be interesting theoretical reasons. In the end, the scaling function is again of the form (3.3) with α still the same as in the NCOP case and given by (4.8). However, in this case the coefficient β_2 is nonzero in order to satisfy a nontrivial set of boundary conditions obeyed by the scaling function:

$$\beta_2=\begin{cases}-0.0778 , & d=2 , \\ -0.0356 , & d=3 ,\end{cases} \tag{4.13}$$

and

$$\beta_3=\frac{1}{2(2d+1)}\left[(d-1)\beta_2^2-\frac{\pi}{6}\right] . \tag{4.14}$$

V. SHORT-DISTANCE BEHAVIOR

The results in the last section assumed that one works in the distance regime $\xi \ll R \ll L(t)$. The theory is also capable of treating the short-distance behavior. Consider the short-distance region $R \le \xi \ll L$, in this case the theory gives the analytic result

$$C_\Psi(\mathbf{R},t)=\Psi_0^2\left[1-\frac{W(\mathbf{R})}{L}+O\left[\frac{1}{L^2}\right]\right] , \tag{5.1}$$

where

$$W(\mathbf{R})=\frac{\xi}{\sqrt{2}}\left[1+\frac{R^2\kappa_1}{4d}+\cdots\right] \tag{5.2}$$

and

$$\kappa_1=\frac{1}{\Psi_0^2}\int_{-\infty}^{+\infty}\frac{dx}{\xi}\left[\frac{d\sigma(x)}{dx}\right]^2 \tag{5.3}$$

is proportional to the surface tension for the system. Thus, the short-distance behavior of the correlation function $C_\Psi(\mathbf{R},t)$ are governed by detailed properties of the interface which are characterized by the value of the order parameter, the interfacial width, and the surface tension. In this regime, $C_\Psi(\mathbf{R},t)$ is a smooth analytic function of $\mathbf{R}$. This result holds for both the NCOP and COP cases.

The theory can treat the entire $\mathbf{R}$ range and as $\mathbf{R}$ increases one finds for $\xi \ll R \ll L$ that (5.1) still holds but W has crossed over from (5.2) to

$$W(\mathbf{R})=\alpha|\mathbf{R}| , \tag{5.4}$$

where α is the same α given by (4.8) and one matches onto the scaling form (3.3).

VI. LARGE-x LIMIT

While the emphasis in this paper has been on the small-x behavior of the scaling function, one can also work out the large-x behavior. It is, however, probably more difficult to measure this behavior accurately experimentally since it is in this region where $F(x)$ is small. The result of the theory in Refs. 99 and 100 is that $F(x)$ can be determined analytically for large x. For a NCOP system one has a Gaussian form

$$F(x)=F_0 x^{-(d-\pi/2\mu)}e^{-(\mu/2)x^2} , \tag{6.1}$$

where μ is given by (4.11) and F_0 is a constant that has been determined numerically. For the COP case one obtains the more involved oscillatory form

$$F=F_0x^{-2d/3}\exp\left[-\frac{3}{8\nu^{1/3}}\left\{x^{4/3}-\frac{\pi}{3\nu^{2/3}}x^{2/3}\right\}\right]\times\cos\left[\sqrt{3}\tfrac{3}{8}\nu^{1/3}\left\{x^{4/3}+\frac{\pi}{3\nu^{2/3}}x^{2/3}+\phi\right\}\right]\,, \tag{6.2}$$

where F_0 and ϕ are constants which have been determined numerically and, as in the NCOP case, the final results depend only on d with

$$\nu=\begin{cases}0.3376\ , & d=2\ ,\\ 0.1752\ , & d=3\ .\end{cases} \tag{6.3}$$

VII. PERTURBATIONS

The theoretical results described above are for the specific class of models defined by (4.1). There are a number of possible perturbations which might change the scaling properties and the coefficients α and β_i. Since most of these possible changes are, as yet, unexplored theoretically, I can only raise a warning signal that these effects may change the scaling function.

A. Thermal effects

In the analysis above it was assumed that the quench was to zero temperature. If the quench is to a nonzero temperature T_F, then one can ask how the results for the scaling function F are modified. One expects[117] that thermal effects will have the effect of renormalizing only the equilibrium properties like the "magnetization" Ψ_0 which goes to $\Psi_0(T_F)$ and the interfacial profile $\sigma(z)$ and the interfacial width ξ will now depend on T_F. However, as long as $R \gg \xi(T_F)$, one expects the coefficients β_i and α to be independent of T_F.

B. Off-critical quenches

The discussion of the model given by (4.1) up to this point has been limited to the case of critical quenches where, by virtue of symmetry, the average value of the order parameter is zero,

$$\langle\Psi\rangle=0\ . \tag{7.1}$$

The case of off-critical quenches for the case of a COP is of great interest since spinodal decomposition for an alloy mixture will typically not be for a 50-50 composition. In this case one has that

$$\langle\Psi\rangle=M \tag{7.2}$$

and M has a fixed time-independent value. One then has the problem of treating (4.1) with the constraint (7.2). As one changes M from 0 to Ψ_0, one changes the late-time morphological structure from the ramified structure[121,122] to a structure of compact objects (spheres) coarsening in a background of the majority phase. The theory of a dilute set of such compact objects is given by the Lifshitz-Slyozov[76] theory and the growth law is $t^{1/3}$ for all values of M. It is clear that F does depend on M, but theoretical results in the literature[123,124] do not make strong statements about the variation of F with M. In the simulation work of Rogers and Desai[118] they find a rather weak dependence of F on M for M ranging from 0 to 0.4. The main change they find is a lowering of $F/(1-M^2)$ for small x for the $M=0.4$ case relative to the $M=0$ case. They claim that this may be a nonscaling effect which may diminish for longer times. It is worth noting that, for $M=0.4$, the system has already crossed over to a final morphology of compact structures embedded in a majority phase matrix.

The extension of the theory in Ref. 100 to the $M\neq0$ case is relatively straightforward and involves reformulating the theory such that the probability distribution governing the auxiliary field m, appearing in $\sigma[m]$, allows for a nonzero value of m. This complicates the theory, but not in a prohibitive way. It is clear that the theory can be developed in much the same way as for $M=0$ and that the growth law will still be given by $L\sim t^{1/3}$. It is clear that the scaling function F and the α and β coefficients do depend on M. A preliminary analysis shows a lowering of F for small x in agreement with Ref. 121. Complete theoretical analysis will be available in the near future.

C. Strain effects and solid symmetries

For an ordering solid whose elastic properties are not approximately isotropic, the morphology of the growth structure will be anisotropic[125–129] and this anisotropy will be reflected in the coefficients α and β_i. The specific nature of this dependence is not yet known but under current theoretical investigation.

D. Role of impurities

If there are quenched impurities in the system then they have a dramatic effect ont he growth kinetics in the system. The growth law can be changed[77–85] to a logarithmic form and, indeed, ordering may be stopped at some point. The nature of scaling in ordering systems with quenched disorder is not understood at this point.

E. Multiplicity of order parameter

It is known from the work of Lai[130] that the scaling properties may be influenced by the existence of a multiplicity of competing order parameters which may introduce a competition between different types of interfaces. The model given by (4.1) and (4.2) shows only one type of interface. Systems like Cu_3Au (Refs. 131 and 132) show two different types of interfaces and this appears to influence the scaling properties of such systems. Experimentally one finds anisotropy in the scaling properties of Cu_3Au which will be reflected in the coefficients C_n. Detailed calculations need to be done to determine the modification of the C_n's due to this anisotropy.

F. Flow

It is known[75] that flow in fluid systems and the associated hydrodynamical couplings change the growth law from $t^{1/3}$ to t. It is expected[133] that there will be associated crossover effects in the scaling function.

G. Polymers

It has been proposed[134] that the models (4.1) and (4.2) can be used to describe phase separation in polymer mixtures if $p=1$, V is some variant of the Flory-Huggins free energy, and c is taken to be Ψ dependent. Preliminary calculations seems to indicate that the scaling function and the β coefficients are sensitive to the Ψ dependence of c. This somewhat surprising result must be investigated further.

VIII. CONCLUSIONS

A direct experimental or numerical determination of the coefficients defined by (3.2) and (3.3) can be very useful in elucidating the degree of universality in growth-kinetics problems. For the theoretical model defined by (4.1) there are definite theoretical predictions which can be tested. If we define the ratios

$$R_n = \lim_{t\to\infty} \frac{C_n C_0^{n-1}}{C_1^n} , \tag{8.1}$$

where the C_n are defined by (2.1) in the spatial regime $\xi \ll R \ll L$, then the R_n have universal values for the class of models defined by (4.1). Again, these results do not depend on the form of the potential, the type of underlying lattice, or the nature of the initial conditions. The values of R_n for $n=2$ and 3, $d=2$ and 3, and for conserved and nonconserved order parameter are given in Table I. There are clear differences between the COP and NCOP cases.

TABLE I. The "universal" ratios R_n defined by (8.1) for $d=2$ and 3 for conserved (COP) and nonconserved (NCOP) order parameters.

	$d=2$		$d=3$	
n	NCOP	COP	NCOP	COP
2	0	0.0975	0	0.0631
3	−0.2557	−0.0813	−0.2504	−0.1169

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