Computationally Efficient Modeling of Ordering of Quenched Phases

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(Received 14 July 1986)

Computationally efficient discrete space-time models of phase-ordering dynamics of thermodynamically unstable systems (e.g., spinodal decomposition) are proposed. Two-dimensional lattice (100×100) simulations were performed to obtain scaled form factors.

PACS numbers: 81.30.Hd, 02.70.+d, 64.60.Cn

One of the difficult outstanding problems in phase transitions is that of the ordering dynamics of thermodynamically unstable systems, e.g., quenched binary alloys.[1] The process depends crucially on whether the order parameter of the system is conserved or not. In the former case the process has been called spinodal decomposition. In this Letter, to cover both cases, we use the term *phase ordering* for the ordering process of unstable phases in general.

The purpose of the present Letter is to propose computationally efficient models of phase-ordering dynamics utilizing discretized space and time corresponding to the usual coarse-grained description of the dynamics.

The theoretical study of phase ordering has a long history since the days of Cahn and Hilliard,[2] but the true revitalization of the study came from the observation of the approximate scaling law in Monte Carlo simulations by Marro *et al.*[3,4] They suggested that the normalized form factor $S(\mathbf{k},t)$ has a scaling regime in which it behaves as

$$S(\mathbf{k},t)=l(t)^d\Phi(l(t)k), \tag{1}$$

where $\mathbf{k}$ is the wave vector, t the time, Φ a master function (scaling function), $l(t)$ a time-dependent length scale which behaves as $l(t)\sim t^{\phi}$ for some positive number ϕ, and d the spatial dimensionality. Furukawa[5] also argued for (1).

Experimentally, the exponent seems to behave as follows. When the order parameter is not conserved $\phi\simeq\frac{1}{2}$,[6] and when it is conserved but without hydrodynamic interactions, $\phi\simeq\frac{1}{3}$.[7] If hydrodynamics is relevant, $\phi\simeq1$.[8] Analytically, Ohta, Jasnow, and Kawasaki[9] studied the nonconserved-order-parameter (NOP) case and obtained ϕ as well as Φ. For the conserved-order-parameter (COP) case, Ohta[10] studied the case of the off-critical quench and obtained $\phi=\frac{1}{3}$ as well as Φ (see also the work of Kawasaki and Ohta[11]). Although for initial stages a successful theory exists,[12] there is no reliable theoretical work for the scaling regime in the most interesting case of critical quenching.

Extant Monte Carlo simulations are without hydrodynamic interactions because of their long-range nature. The NOP case has been studied by Phani *et al.*[4] They found $\phi\simeq\frac{1}{2}$ and obtained Φ. For the COP case, extensive simulations have been performed by Marro *et al.*[3] They found that $\phi\sim0.2$ to 0.3 depending on the depths of quenching and the off criticality. It is probable that these simulations are not for sufficiently long time.[13] Petschek and Metiu and Miyazaki *et al.*[14] numerically solved the Cahn-Hilliard (CH) equation but they did not study the scaling regime.

It is clear that we need computationally efficient methods to study the scaling regime. To this end the most effective way is to construct computationally efficient minimal models of phase ordering which capture the essence of the physics.

Let us first recall that the conventional description of phase ordering uses spatially coarse-grained free-energy functionals. This description is inevitably coarse-grained in time also; the time-dependent Ginzburg-Landau and CH equations should be regarded as *Ansätze* connecting the coarse-grained free energy with the dynamics.

We propose that the space-time coarse-graining in the conventional models should be explicitly imposed by use of a discretized space-time lattice. The spatial lattice describes the dynamics of regions of order not much smaller than the correlation length ξ. Notice that, if we discretize existing partial differential equation models using standard schemes, the single-cell dynamics, which must be purely relaxational, becomes oscillatory or even chaotic when we choose the time increment to be big, viz. of order ξ^2/D, where D is the typical diffusion coefficient. Therefore, we should model phase-ordering dynamics directly without referring to any continuum model. This is tantamount to proposing new *Ansätze*.

First, we mimic the single-cell behavior by a one-to-one map on the set of real numbers R (identified with the set of possible values of the order parameter), and then we couple them spatially through local averaging. The conservation of the order parameter is the property of the relation among spatially coupled cells. Hence, to mimic the behavior of a single cell, we may ignore the conservation law. Thus, the purely relaxational single-cell behavior can be mimicked by a one-to-one map on R with two hyperbolic sinks and one hyperbolic source. The former corresponds to new equilibrium order-parameter values after quenching, and the latter the free-energy minimum state before quenching. Figure 1

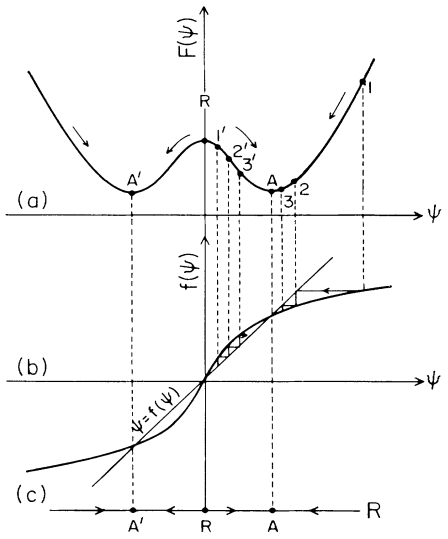

FIG. 1. (a) The coarse-grained free energy F at roughly the scale of the correlation length. If the local order parameter takes the value at 1 (1'), then the next value after a unit time step is given by the value at 2 (2'), etc. This motivates the one-to-one $f:R \to R$ as is shown in (b) which can describe the behavior. A and A' are hyperbolic sinks and R the source. (c) The flow due to this map. What we really need is only (c) which requires a one-to-one map similar to (b).

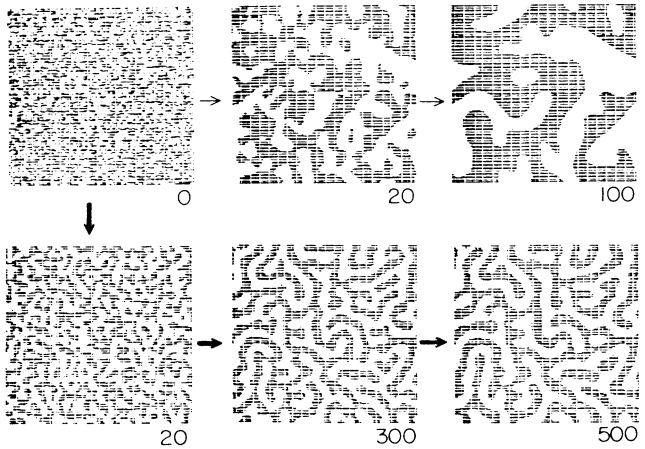

FIG. 2. Thin arrows indicate the time evolution of textures of the nonconserved-order-parameter case, and thick arrows that of the conserved-order-parameter case, both from the same initial random configuration 0. The numbers denote necessary time steps from 0. Only cells with positive order parameter are marked.

and its legend *motivate* the choice of the map; we must stress that this is *not* the derivative of the map. Strictly speaking, in our *Ansätze* we do not even need free-energy functionals; only the features of local dynamics [Fig. 1(c)] are needed. We believe that any such one-to-one map $f(x)$ is in the same universality class. It is one of the critical points of our modeling that the one-to-one nature of the map automatically excludes[15] the possibility of oscillatory or chaotic local dynamics which plagues the large-time-step discretization of continuum models.

Thus, we mimic the single-cell discrete dynamics as

$$\Psi(t+1,n)=f(\Psi(t,n)), \tag{2}$$

where $\Psi(t,n)$ is the value of the order parameter in the cell n at time t. Next, we must introduce the intercell coupling. Without the conservation of the order parameter the diffusion should cause the increment of Ψ proportional to its difference from the average of the order parameter in the neighborhood cells. Thus the discrete model for the NOP cases reads[16]

$$\Psi(t+1,n)=f(\Psi(t,n))+D[\langle\langle\Psi(t,n)\rangle\rangle-\Psi(t,n)]\equiv\mathcal{F}[\Psi(t,n)], \tag{3}$$

where $\langle\langle\rangle\rangle$ is the average in the neighborhood except for the center cell (the nth cell). We believe that any isotropic local average is acceptable. Here we define $\langle\langle\rangle\rangle$ on the square lattice as follows:

$$\langle\langle\Psi(t,n)\rangle\rangle=\sum(\Psi \text{ in the nearest-neighbor cells})/6+\sum(\Psi \text{ in the next-nearest-neighbor cells})/12. \tag{4}$$

The conservation of the order parameter implies the local sum rule that when there is an exchange of order-parameter values between a cell and its neighboring cells, there should not be any net change of the order parameter inside of the neighborhood surrounding the center cell. Since the net gain of the order parameter by the center cell is given by $\mathcal{F}[\Psi]-\Psi$, the discrete model for the conserved case reads

$$\Psi(t+1,n)=\mathcal{F}[\Psi(t,n)]-\langle\langle\mathcal{F}[\Psi(t,n)]-\Psi(t,n)\rangle\rangle, \tag{5}$$

where the subtraction corresponds to the extra Laplacian in the CH equation.

The models constructed above are deterministic models. It is generally believed that in the scaling regime stochastic effects are not important.[17] Thus for our preliminary study we use deterministic models in accordance with recent theoretical attempts.[9,11] We are studying models which include noise, with no particular loss of computational efficiency.[18] Our deterministic models may be regarded as coupled map-lattice models.[19] We believe that computationally efficient space-time discrete modeling of real nonequilibrium phenomena[20] is worth persuing seriously.

In our actual simulations, we adopted several maps in-

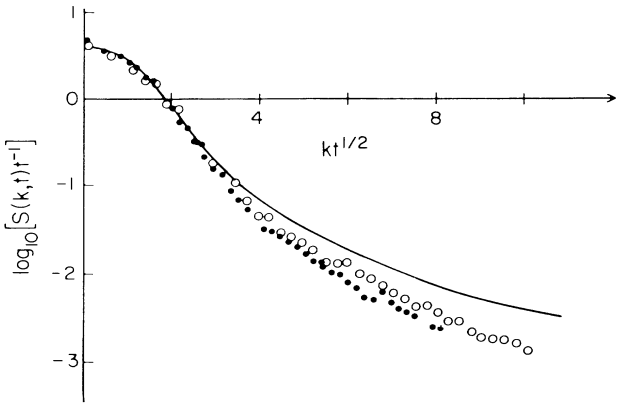

FIG. 3. The scaled form factor Φ for the nonconserved-order-parameter case. Filled circles denote Φ at $t=100$, and open circles at $t=180$. The curve is the theoretical curve by Ohta *et al.* (Ref. 9) shifted and scaled appropriately. For smaller $kt^{1/2}$ we have a reasonable master curve which is in agreement with the theoretical curve. For larger $kt^{1/2}$ we have not yet obtained a single master curve, but we see an increase of the tail toward the theoretical curve.

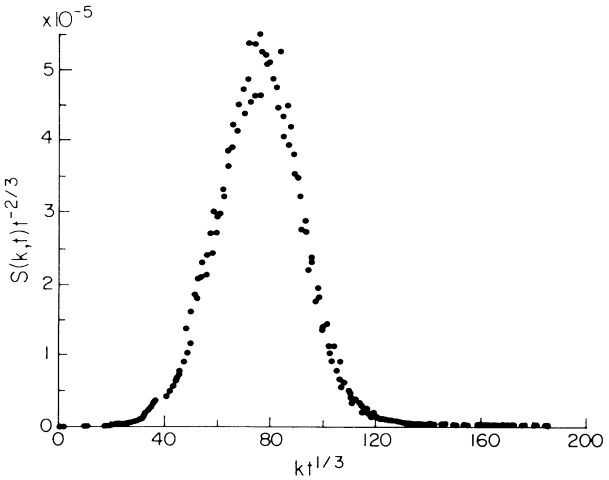

FIG. 4. The scaled form factor Φ for the conserved-order-parameter case. The results for $t=500$, 550, 600, 650, and 700 are superposed. We can include the data for t larger than 300 with slightly more scatter of points. In the vicinity of the peak, sampling error cannot be ignored.

cluding piecewise linear maps and $f(x)=A\tanh x$. Here we present results due to the hyperbolic tangent model, but outcomes of other maps are very similar, supporting the above-mentioned universality belief, which strongly supports our *Ansätze*. We will not discuss the relation between parameters in our models and those in more "realistic" models but note that our simulations shown here correspond to deep quenching with "hard" boundary walls.[21]

All our results were obtained on the two-lattice of size 100×100 with periodic boundary conditions and with $f(x)=1.3\tanh x$, $D=0.5$. Form factors were calculated as averages over thirty different initial configurations. Calculations were performed on a VAX-11 computer (without array processors). It took 1.98 sec on the central processing unit to update the whole lattice in the NOP case and 3.33 sec on the central processing unit in the COP case. As can be seen from Fig. 2, well-developed patterns appeared within twenty updates in both cases. In this figure the initial conditions are chosen to be random (uniformly distributed between ±0.125).

In Fig. 3, $S(k,t)t^{-1}$ for different times are superposed as functions of $kt^{1/2}$ for the NOP case. From this we may conclude that there is a master function with $l(t)=t^{1/2}$. Actually, what we did is to plot $S(k,t)t^{-2\phi}$ vs kt^{ϕ} for various exponents ϕ and searched the range of ϕ which give reasonable master curves. We found that $\phi=0.49\pm0.03$ allows reasonable single master curves. The Monte Carlo $S(k,t)^4$ for the larger k region behaves as $k^{-2.5}$, violating Porod's law.[22] In contrast, our results are consistent ($\sim k^{-3}$) with this law [$k^3S(k,t)$ exhibits wide flat plateaus].

In Fig. 4 an analogous master-curve plot for the COP case is shown. This time we found that the exponent $\phi=0.33\pm0.03$ gives reasonable master curves. Unfortunately, in this case there is no analytic result that we can compare with. If we choose $A=1.2$, $D=0.1$, we can simulate the so-called soft-wall case.[23] Although the wall thickness eventually becomes much smaller than the typical size of patterns, the transition from the thick (or soft) to the thin-wall regime takes very long. In this transition regime, we got $\phi\sim\frac{1}{4}$ to have a single master curve. Therefore, we suspect that the exponents so far observed by the Monte Carlo method are not truly in the scaling regime, where the walls must be thin relative to the representative scale of patterns.

To summarize, we have proposed cell-dynamical-system models (or *Ansätze)* of phase-ordering dynamics, which are sufficiently realistic and computationally efficient.

We are grateful to T. Ohta and Y. Shiwa for teaching us the present status of the theory; especially, Ohta's lectures were highly useful. We gratefully acknowledge Ohta's critical reading of the manuscript and N. Goldenfeld's useful comments and help in revising the manuscript. We are also grateful to M. Salamon for his interest in and generous support for the present project and to Mrs. V. Metze for her facilitating our simulation. The work is supported, in part, by National Science Foundation Grant No. DMR-84-15063 and Materials Research Laboratory Grant No. NSF DMR-83-16981.

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$$\mu(t+\Delta t,x) = (1-2dD')F_{\Delta t}(\Psi(t,x)) + dD'\langle\langle F_{\Delta t}(\Psi(t,x))\rangle\rangle.$$

Equation (3) is essentially obtained by identification of F_1 with f. This suggests how to construct f from g. We can do a similar thing for the CH equation.

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