PHYSICAL REVIEW E VOLUME 52, NUMBER 4 OCTOBER 1995

Domain growth in computer simulations of segregating two-dimensional binary fluids

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(Received 2 May 1995)

We studied phase segregation kinetics with hydrodynamic interactions, following a quench, in the two-dimensional binary fluid lattice gas model of Rothman and Keller [J. Stat. Phys. **52**, 1119 (1988)]. Carrying out computer simulations at different overall fluid densities d, with equal volume fractions of the two components, we find that the growth of domain sizes $R(t)$ at different d has a scaling behavior with all data well fitted by $R(t)/R_s = a + b(t/t_s)^{2/3}$. The characteristic lengths $R_s(d)$ and times $t_s(d)$ are related in a simple way to the viscosity and surface tension of the system at different values of d. We also discuss the growth exponents expected in the general case of phase segregation with hydrodynamic interactions.

PACS number(s): 64.70.Ja, 61.20.Ja, 64.60.Qb, 64.75.+g

I. INTRODUCTION

Phase ordering is the process through which a system evolves towards equilibrium following a temperature quench from a homogeneous phase into a two-phase region. The theoretical understanding of this highly nonlinear phenomenon remains, after more than three decades since the classical work of Cahn and Hilliard [1], a very challenging problem. During this period there has been an accumulation of much experimental and computer simulation evidence showing that the domain growth in phase separating systems is a scaling phenomenon [2–5]. More precisely, there exists at late times a single characteristic length $R(t)$, such that the domain structure is in a statistical sense independent of time when lengths are scaled by $R(t)$, the average domain size at time t. The dependence of $R(t)$ on time, the growth law, appears to have the asymptotic form $R(t) \propto t^{\alpha}$, with the growth exponent α depending only on the conservation laws obeyed, the dimensionality of the system, and the symmetry of the order parameter [6].

Much effort has gone into trying to predict or justify the growth exponents characterizing different classes of systems, but success is still limited. For systems with conserved order parameter and without momentum conservation, such as binary alloys, Lifshitz and Slyozov found in their pioneering work [7] that for small volume fractions of one of the species the domain growth should follow a power law with exponent $\alpha = \frac{1}{3}$. This result has been confirmed by experiments and computer simulations [2]. Although the first theoretical and computational attempts [8] to extend it to critical quenches (equal volume fractions of the two species) were not very successful, its validity in diffusive systems for all concentrations and for all dimensions is now universally accepted [2,9]. This is indeed a remarkable universality, reminiscent of the $t^{\frac{1}{2}}$ diffusive behavior outside the coexistence region.

The situation is much less clear, both theoretically and experimentally (including computer simulations), about phase segregation with hydrodynamic interactions, i.e., when both the order parameter and momentum are locally conserved, as in binary fluid mixtures following a quench. This uncertainty concerns both the correct coupling between the Cahn-Hilliard equation for the order parameter and the Navier-Stokes equation for the fluid velocity as well as the growth exponent α. Based mainly on dimensional analysis, Siggia predicted [10] that the domain growth in three-dimensional (3D) binary fluids at equal concentrations (critical quench) should have a crossover, as time goes on, from an $\alpha = \frac{1}{3}$ regime to an $\alpha = 1$ regime. This crossover was indeed observed in experiments [11–13]. The linear growth was also confirmed by some numerical simulations [14–16], but not others [17,18], despite a very careful analysis [17]. Much work remains to be done to understand the physical mechanisms driving domain growth at different volume fractions, and to improve the very demanding computational methods in 3D.

Siggia's derivation of a linear growth was analyzed in 2D by San Miguel *et al.* [19], who argued, using linear stability analysis, that the $\alpha = 1$ regime is not possible in 2D. For 2D binary fluid mixtures with approximately equal volume fractions they proposed droplet coalescence to be relevant in domain growth; this mechanism gives, by simple dimensional analysis, an exponent $\alpha = \frac{1}{2}$ in 2D and $\alpha = \frac{1}{3}$ in 3D. (Droplet coalescence, rather than the Lifshitz-Slyozov mechanism, is probably responsible for the $\alpha = \frac{1}{3}$ regime observed in 3D binary fluids [2,10,11,13,20,21].) An $\alpha = \frac{1}{2}$ exponent was observed in molecular dynamics (MD) simulations on a 2D Widom-Rowlinson type mixture for early times by Velasco and Toxvaerd [22], who also noted a rise in slope at latter times. Subsequently, Ossadnik *et al.* [23] argued on the basis of more extensive MD simulations that the exponent should stay at $\alpha = \frac{1}{2}$. A similar result was obtained recently by Leptoukh *et al.* [24]. On the other hand lattice Boltzmann (LB) simulations in 2D yield a $\frac{2}{3}$ exponent [14,16], as predicted by Furukawa [25,26], while Langevin dynamics simulations find an exponent close to $\frac{2}{3}$ [27,28]. To resolve this question we carried out simulations on a 2D binary fluid immiscible lattice gas model (ILG) of Rothman and Keller [29] (see also Ref. [30] for a related model), where we could gather reliable statistics. The model is described in Sec. II and the results in Sec. III. A description of our current understanding of different growth regimes in binary fluids is given in Sec. IV, where we also discuss possible additional simulations and experiments.

II. DESCRIPTION OF MODEL

The 2D ILG is based on the Frisch, Hasslacher, and Pomeau (FHP) model [31], which is a discrete — in both space and time — lattice gas model on a triangular lattice. In the version of FHP used by Rothman and Keller up to seven particles may reside at a site. At each site there are seven available velocity states: six unit velocity states connecting neighboring sites on the lattice and a zero velocity state corresponding to a "rest particle." The discrete time dynamics consists of two steps. First, particles at one site change their velocities according to collision rules conserving the mass (particle number) and linear momentum. Then they travel to the neighboring sites according to their new velocities and the process repeats itself at the next time step. The macroscopic behavior of this model is described at low velocities by incompressible Navier-Stokes-like equations [32].

The ILG is built on the above foundation. Two kinds of particles are introduced, "red" and "blue," and the collision rules are changed to encourage phase (color) segregation while conserving momentum and the number of reds and blues. This is accomplished by introducing a red field $r(\vec{x})$ and a blue field $b(\vec{x})$. The red field is defined as the set of seven Boolean variables

$$r(\vec{x}) \equiv \{r_i(\vec{x}) \in \{0,1\},\ i = 0,1,...,6\}; \tag{1}$$

$r_i(\vec{x})$ indicates the presence or absence of a red particle with velocity $\vec{c}_i$ at lattice site $\vec{x}$; $\vec{c}_0 = \vec{0}$ and $\vec{c}_i$ through $\vec{c}_6$ are unit vectors connecting neighboring sites on the triangular lattice. The blue field is defined in a similar fashion. Red and blue particles may simultaneously occupy the same site but not with the same velocity. The phase segregation is generated by allowing particles from nearest neighbors of site $\vec{x}$ to influence the output of a collision at $\vec{x}$. Specifically, a local color flux $\vec{q}[r(\vec{x}), b(\vec{x})]$ is defined as the difference between the net red momentum and the net blue momentum at site $\vec{x}$:

$$\vec{q}[r(\vec{x}), b(\vec{x})] = \sum_i \vec{c}_i[r_i(\vec{x}) - b_i(\vec{x})] \tag{2}$$

and a local color field $\vec{f}(\vec{x})$ as the lattice gradient of the order parameter field $\Phi(\vec{x})$:

$$\Phi(\vec{x}) = \sum_i [r_i(\vec{x}) - b_i(\vec{x})] \ , \tag{3}$$

$$\vec{f}(\vec{x}) = \sum_i \vec{c}_i \Phi(\vec{x} + \vec{c}_i) \ . \tag{4}$$

The "work" performed by the color flux against the color field is

$$W(r, b) = -\vec{f} \cdot \vec{q}(r, b) \ . \tag{5}$$

The result of a collision at site $\vec{x}$, $r \to r'$ and $b \to b'$, is then with equal probability any of the outcomes which minimize the "work"

$$W(r', b') = \min_{r'', b''} W(r'', b''), \tag{6}$$

subject to mass, color, and momentum conservation. (This implies a very deep temperature quench, arguably one at $T = 0$.) When implementing the above algorithm we used the scheme proposed originally by Rothman and Keller which reduces the size of the collision table by allowing only 36 directions for the color field $\vec{f}$ [29]. This model has been shown to exhibit surface tension and to satisfy Laplace's law [29,33,34].

III. SIMULATION RESULTS

We used a 256 × 256 lattice with periodic boundary conditions and studied the evolution of the system at seven reduced densities d: 0.29, 0.3, 0.31, 0.35, 0.4, 0.45, and 0.75, where $d = \frac{\rho}{7}$ and ρ is the average number of particles per site. The reason for using these densities is that for d smaller than about 0.25 the segregation no longer takes place [33–35]. Also, the adherence of the FHP model to the Navier-Stokes equations is true only for $d < 0.5$ [32].

At each density the initial distribution of particles was random, with reds and blues equally probable (critical quench). A typical evolution of the two-fluid mixture is shown in Fig. 1. To probe the domain structure we studied the pair correlation function at time t following the quench:

$$C(\vec{r}, t) = \frac{1}{\mathcal{V}} \left\langle \sum_{\vec{x}} \Phi(\vec{x}, t) \Phi(\vec{x} + \vec{r}, t) \right\rangle \ , \tag{7}$$

where $\mathcal{V}$ is the volume, and its Fourier transform, the equal time structure factor:

$$S(\vec{k}, t) = \langle |\Phi(\vec{k}, t)|^2 \rangle, \tag{8}$$

the average being in both cases over the initial conditions. The spherical (circular in 2D) averages of the above provide $C(r,t)$ and $S(k,t)$. The late time scaling forms for $C(r,t)$ and $S(k,t)$ are [2,5]

$$C(r, t) = G(r/R(t)) \ , \tag{9}$$

$$S(k, t) = R(t)^D F(kR(t)), \tag{10}$$

where D is the spatial dimensionality and G and F are time independent functions. $F(u)$, $u = kR(t)$, is the structure factor and its tail should satisfy *Porod's law*: $F(u) \propto u^{-(D+1)}$, $u \gg 1$. As a measure of the domain size we made the usual choice, the dis-

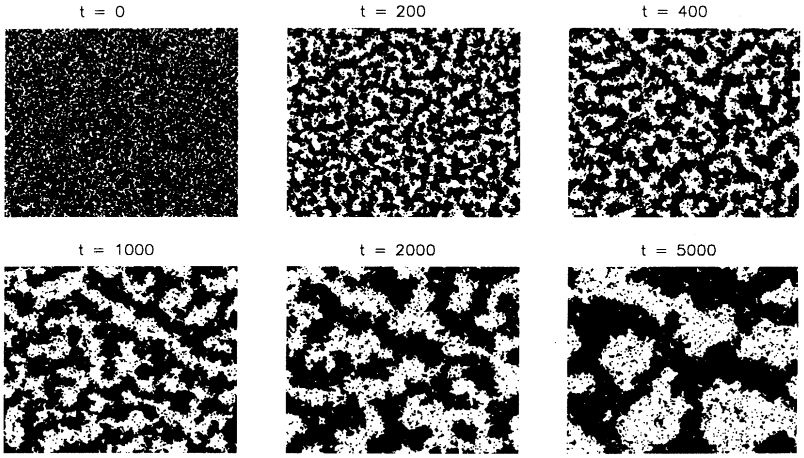

FIG. 1. Time evolution of the two-fluid mixture for $d = 0.3$. The sites with positive order parameter are black; if the order parameter is zero the site is colored by looking at the nearest neighbors or randomly (in this order).

tance at which $C(r,t)$ first crosses zero. As a consistency check we computed also the average wave number $\overline{k} = \sum_k kS(k,t)/\sum_k S(k,t)$; its inverse should also be a measure of the domain size [2]. For each density the average of at least 16 independent runs was taken to determine the growth law $R(t)$. The total number of runs for all densities was 148.

The dependence of the domain size on time is shown in Fig. 2 for all densities. The collapse of the curves into a single one through scaling in both length and time is possible for all reduced densities d smaller than 0.5. (For $d = 0.75$ the scaling is not possible, although the asymptotic exponent seems to be also $\frac{2}{3}$, as for the other curves; see Fig. 2.) We found the best scaling (up to a multiplicative constant in both length and time) and the result is shown in Fig. 3. This shows both that the scaling is very good and that the resulting function is $r = a + b\tau^{\frac{2}{3}}$, where $r = \frac{R}{R_s}$ and $\tau = \frac{t}{t_s}$. This is in agreement with LB simulations of a similar 2D binary fluid [14] and with the results in [25–27]. The characteristic length and time scales R_s and t_s are related to the physical parameters of the system as suggested by the derivation of the relation $R(t) \propto t^{\frac{2}{3}}$ [26]. This derivation is based on a dimensional analysis of the Navier-Stokes equation, viewed as a balance between a driving force and a "friction" force. The driving force is the gradient of the surface tension induced pressure of the liquid mixture, while the friction force is made up of viscous and "inertial" parts. When the viscous friction dominates a linear growth regime is predicted as in Siggia [10]. As the domains become large we get to an inertia dominated regime with growth exponent $\alpha = \frac{2}{3}$. The crossover between these two regimes takes place [5,25] when the domain size is of the order

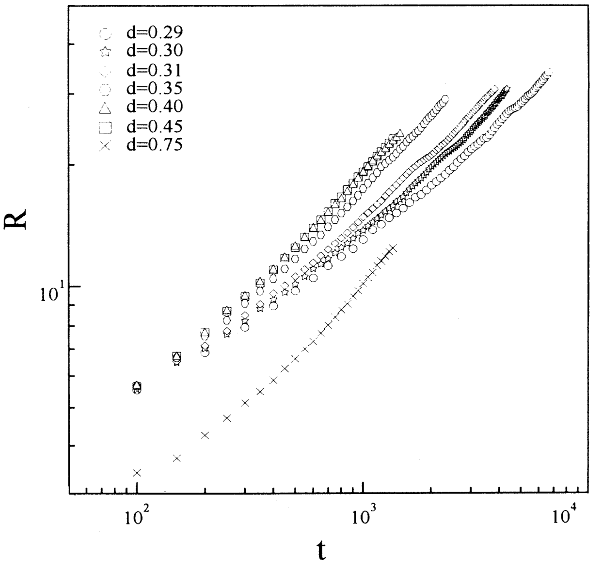

FIG. 2. Domain size (in lattice units) as a function of time for all densities; log-log plot. The $d = 0.75$ curve has been moved down 0.125 log units for easier viewing.

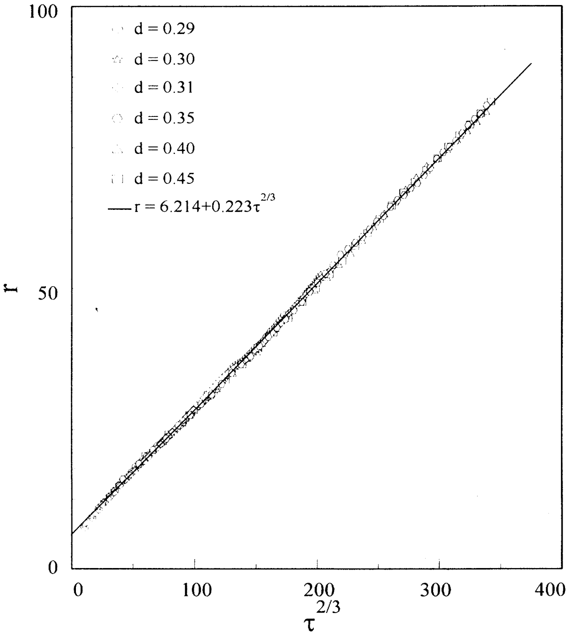

FIG. 3. Domain size as a function of time for all densities $d < 0.5$, in scaled variables r and $\tau^{\frac{2}{3}}$; $r = \frac{R}{R_s}$ and $\tau = \frac{t}{t_s}$. The best line fit is drawn.

of the hydrodynamical length $R_h = \frac{\nu^2\rho}{\sigma}$, where ρ is the density, ν is the kinematic viscosity, and σ is the surface tension coefficient. The growth law is then given by $\frac{R(t)}{R_h} \propto (\frac{t}{t_h})^{\frac{2}{3}}$, $\frac{R}{R_h} \gg 1$, where $t_h = \frac{\nu^3\rho^2}{\sigma^2}$.

In order to estimate R_h and t_h and see how they compare with R_s and t_s for the FHP model, which behaves like a fluid in the continuum limit [32], we need to know ν and σ; the density ρ is simply the average number of particles per site. The kinematic viscosity ν was computed as a function of density in the Boltzmann approximation and also through computer simulations [32,34,36,37]. The 2D ILG was shown to exhibit surface tension and to satisfy Laplace's law, and the surface tension coefficient σ was determined using the Boltzmann approximation and checked through simulations [33,34]. Using these results we estimated R_h and t_h; we show them in Fig. 4 together with R_s and t_s as found by collapsing the growth laws into a single curve. (As R_s and t_s were both determined up to a multiplicative constant, the best fit was used.) The agreement is good considering the approximations involved when determining ν and σ and the g factor problem of the FHP models [32]. This indicates that the underlying similarity of the growth laws at different densities of this model is indeed related to the physical parameters ρ, ν, and σ through R_h and t_h.

In Fig. 5 we show the structure factor $F(u)$, with the maximum normalized to 1, for all densities. While it seems to be time independent at late times, as expected if dynamic scaling is to be true, Porod's law is best satisfied for the higher densities. This is probably a consequence of the discreteness of the model. The small u behavior, $F(u) \approx u^2$, is in agreement with [14]. Some very recent MD simulations involving quenching below the triple point of a Lennard-Jones fluid indicate a scaled structure function with a time dependent prefactor [38]. We do not see such a behavior in our simulation.

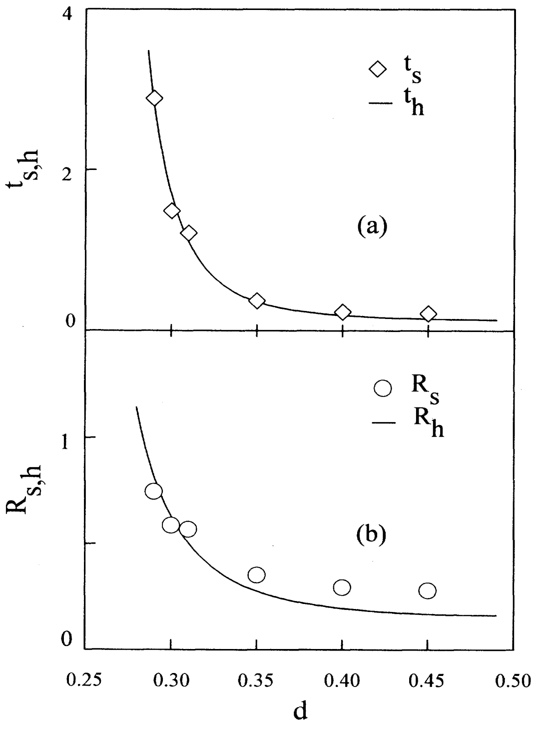

FIG. 4. The scaling t_s and hydrodynamical t_h times (a) and the scaling R_s and hydrodynamical R_h lengths (b) are shown; R_h and t_h are defined in the text.

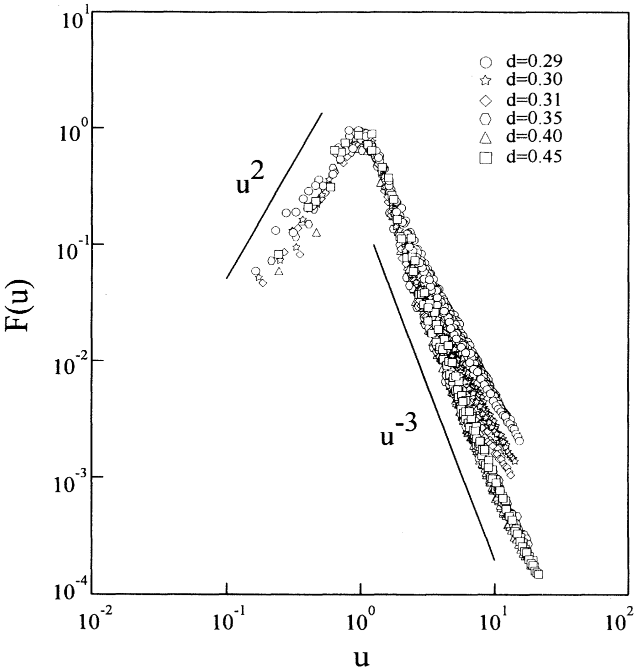

FIG. 5. The structure factor $F(u)$ for all densities. For each density $F(u)$ is shown beginning at $t = 1000$ in steps of 1000, up to the latest available time.

IV. DISCUSSION

We now summarize briefly the picture that seems to be emerging for the growth exponents of binary fluids at close to equal volume fractions of the two components, in both two and three dimensions. This picture appears to us consistent with all the theoretical, computational, and experimental work mentioned in this paper. At very late times (or large domain sizes $R > R_h$) the domain growth is dominated by inertial hydrodynamics which gives an $\alpha = \frac{2}{3}$ exponent in both 2D and 3D. This regime is preceded in 3D ($R < R_h$) by a viscous hydrodynamic regime with $\alpha = 1$, and at even earlier times by a $t^{\frac{1}{3}}$ growth, driven by Brownian droplet coalescence, as observed in [10–13]. In 2D the linear growth was ruled out on linear stability grounds by San Miguel *et al.* [19]; therefore the inertial hydrodynamic regime should be preceded directly by growth through droplet coalescence which gives $\alpha = \frac{1}{2}$.

This scenario is clearly consistent with both our and Ref. [14] simulation work in 2D which are in the $R(t) > R_h$ regime, and it could explain the $\frac{1}{2}$ exponent found in

[22] and [24]: probably $R(t) < R_h$. It is also consistent with the experimental and simulation work in 3D [11–14]. In all these cases R_h can be estimated to be bigger than the maximum domain size shown. On the other hand the $\alpha = 1$ regime is never observed in [18], where the 3D ILG is used, although the system size is the same as in [14]. Using values of η and σ as given in [39], R_h can be estimated to be less than 1, so $R > R_h$ at all times; the exponent seems indeed to be around $\frac{2}{3}$.

To check this picture further it would be interesting to carry out more simulations in 2D for $R(t) < R_h$ and in 3D for $R(t) > R_h$. This is possible using the lattice Boltzmann models, for which the viscosity and the surface tension coefficient can be varied independent of the density [14]. If our analysis is correct, the growth in the region $R(t) < R_h$ should yield in 2D $\alpha = \frac{1}{2}$ while for 3D there should be a crossover to a $t^{\frac{2}{3}}$ regime when $R > R_h$. The experimental study of a crossover to $\alpha = \frac{2}{3}$ is much more difficult. The reason is that the time scales for spinodal decomposition are typically very small for fluids [2]. This difficulty is usually overcome by making measurements very close to the critical point, a typical temperature quench being of the order of 10–100 mK. As σ vanishes at $T = T_c$, R_h for such quenches is bigger than 0.1 cm, while the typical domain size studied is of the order of 10^{-3} cm.

While our analysis and discussion has been in terms of R_h and t_h, the scaling variables prefered by experimentalists are $R_* = \xi$ and $t_* = \frac{\xi^2}{D}$, where ξ is the correlation length and D is the diffusion constant. Quenches performed at different temperatures have been found to scale in these variables in both the $\alpha = \frac{1}{3}$ [13] and $\alpha = 1$ regimes [11]. When growth is dominated by hydrodynamics ($\alpha = 1$) scaling by R_h and t_h gives $R(t) \propto \frac{\sigma}{\eta}t$, while scaling by R_* and t_* yields $R(t) \propto \frac{D}{\xi}t$. These two are equivalent as $\sigma \propto \frac{k_BT}{\xi^2}$ and $D \propto \frac{k_BT}{\eta\xi}$.

We note finally that away from a critical quench, at volume fractions in the region 0.02–0.3, the droplet coalescence mechanism completely dominates the segregation in 3D, such that only a $t^{\frac{1}{3}}$ regime is observed [13,11,21]. One would expect approximately the same qualitative behavior in 2D, but with a corresponding exponent of $\frac{1}{2}$. At even smaller volume fractions the Lifshitz-Slyozov mechanism might become important, giving an $\alpha = \frac{1}{3}$ exponent in both 2D and 3D.

ACKNOWLEDGMENTS

We would like to thank Frank Alexander, Ricardo Brito, Oliver Penrose, and Vadim Nikolayev for useful discussions. This research was supported in part by NSF Grant No. NSF-DMR 92-13424 and NASA Grant No. NAG3-1414.

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