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Temporally linear domain growth in the segregation of binary fluids

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We report the numerical observation of temporally linear domain growth in a phenomenological model of segregating fluids. Our observation is facilitated by the use of computationally efficient cell dynamical system models.

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Much recent attention has focused on the dynamics of segregation of binary mixtures quenched below the mixing temperature, e.g., spinodal decomposition [1]. Specifically, it is now well established that the segregating domains are characterized by a unique, time-dependent length scale $L(t)\sim t^{\phi}$ (where t is time and ϕ is called the growth exponent). When hydrodynamic effects are not relevant (e.g., binary alloys), it is known experimentally [2] and numerically [3] that ϕ has an asymptotic value of 1/3. When hydrodynamic effects are relevant (e.g., binary fluids), it is theoretically argued [4] that $\phi=1$ asymptotically, and this temporally linear domain growth has also been observed experimentally [5]. The theoretical arguments leave many open questions such as the precise time regime and the volume fraction where this temporally linear growth law should hold. Numerical work clarifying such questions would be very desirable. To date, however, there have been no numerical confirmations of this growth law. In this paper, we report the numerical observation of temporally linear domain growth in a phenomenological model of segregating binary fluids. Our simulation is facilitated by the use of computationally efficient cell dynamical system (CDS) models, which have quite successfully elaborated the nature of spinodal decomposition in the case without hydrodynamics [6], and in a variety of other problems involving reaction-diffusion equations [7].

Our phenomenological model for the dynamics of segregating fluids is a variant of the so-called model H [8], which describes the dynamics of binary fluids and has had considerable success in correctly predicting the dynamical critical exponents of binary fluids. As in model H, our model consists of a scalar density (order parameter) coupled to the hydrodynamic velocity field. With appropriate rescaling [9], our equations have the dimensionless form (in three dimensions)

$$\frac{\partial\psi(\mathbf{x},t)}{\partial t}=-\nabla^2[\psi(\mathbf{x},t)-\psi(\mathbf{x},t)^3+\nabla^2\psi(\mathbf{x},t)]-\bar{\alpha}\nabla\cdot[\psi(\mathbf{x},t)\mathbf{J}(\mathbf{x},t)]\,,$$

$$\frac{\partial J_i(\mathbf{x},t)}{\partial t}=\bar{\eta}\nabla^2 J_i(\mathbf{x},t)+\bar{\sigma}\sum_{k=1}^{3}\nabla_i\nabla_k J_k(\mathbf{x},t)+\bar{\alpha}\psi(\mathbf{x},t)\nabla_i[\psi(\mathbf{x},t)-\psi(\mathbf{x},t)^3+\nabla^2\psi(\mathbf{x},t)]\,. \tag{1}$$

In (1), $\psi(\mathbf{x},t)$ and $J_i(\mathbf{x},t)$ $(i=1,2,3)$ are respectively the order parameter and the dimensionless velocity field as a function of dimensionless space $\mathbf{x}$ $[\equiv(x_1,x_2,x_3)]$ and time t. The rescaled parameters in (1) are the coupling constant $\bar{\alpha}$ ($\bar{\alpha}=0$ corresponds to the usual case of binary alloys) and the transport coefficients $\bar{\eta}$ and $\bar{\sigma}$ (which are the viscosities). Before we proceed, some comments on the form of (1) are in order. First, we consider only the deterministic case as we are interested only in the asymptotic behavior, which is not affected by the presence of thermal noise [10]. In the late stages of growth, the presence of noise terms only alters the interfacial profile between domains and this is an irrelevant factor for sufficiently large domain sizes, i.e., at asymptotic times. Second, (1) would reduce to the standard form of model H [8] if we impose the additional constraint $\nabla\cdot\mathbf{J}(\mathbf{x},t)=0$. However, as was pointed out by Farrell and Valls [9], this extra constraint causes numerical complications and we do not impose it.

Farrell and Valls [9] have numerically studied a more complicated version of (1) (with thermal noise and nonlinear convective terms) in two dimensions. In the late stages of their simulation, they find a domain growth law $L(t)\sim t^{0.65}$. However, it is well known that continuum hydrodynamics is not well defined in two dimensions and it is not clear how this would affect numerical results. Furthermore, the simulation of Farrell and Valls [9] is slowed considerably by the presence of the additional terms. We believe that (1) constitutes the minimal model for studying the late stages of spinodal decomposition in fluids and use it to derive a computationally efficient CDS model, which we use in our simulations.

We do not go into the details of CDS modeling here as these are well documented in the literature [6]. Essentially, this procedure can be understood as an unconventional method of discretizing the corresponding partial differential equations, which enables the use of rather coarse mesh sizes without any loss of numerical stability. The CDS model (in three dimensions) we derive from (1) has the form

$$\psi(n,t+1)=\psi(n,t)-\tfrac{1}{6}\Delta_D[A\tanh(\psi(n,t))-\psi(n,t)+(D/6)\Delta_D\psi(n,t)]-\alpha\nabla_D\cdot[\psi(n,t)\mathbf{J}(n,t)]\,,$$

$$J_i(n,t+1)=J_i(n,t)+\eta\Delta_D J_i(n,t)+\sigma\sum_{k=1}^{3}\nabla_{D,i}\nabla_{D,k}J_k(n,t) \tag{2}$$
$$+\alpha\psi(n,t)\nabla_{D,i}[A\tanh(\psi(n,t))-\psi(n,t)+(D/6)\Delta_D\psi(n,t)]\,,$$

where $\psi(n,t)$ is the order parameter at the discrete lattice site n on a simple cubic lattice at discrete time t; and A, D, α, η, and σ are phenomenological parameters. The parameters A and D are respectively measures of the quench depth and the diffusion. The other parameters are analogous to the corresponding parameters in the continuum model (1). In (2), Δ_D is the isotropically discretized Laplacian operator whose action on a function $f(n)$ is defined as $\Delta_D f(n)\equiv 6[\langle\langle f(n)\rangle\rangle - f(n)]$, where the angular brackets refer to the average of $f(n)$ on the sites neighboring n. This average includes the nearest-, next-nearest, and next-next-nearest neighbors in the relative ratio 6:3:2. Also, in (2), ∇_D is the symmetrically discretized gradient operator. The values of the parameters are dictated by the requirements that the scheme be stable and that the results be reasonable [6]. We choose $A=1.5$ and $D=0.5$, which are good values for simulating the case without hydrodynamics [6]. We can associate mesh sizes with these values of A and D by comparing the CDS scheme with the usual Euler discretization scheme for partial differential equations. The corresponding values are $\Delta t=0.5$ and $\Delta x=2.45$. It is worth noting that these mesh sizes are too large for a stable simulation using an Euler discretization of the given equations [3]. In (1), we set $\bar{\eta}=1$ and $\sigma=2$, following Farrell and Valls [9]. The phenomenological parameters η and σ in (2) are then fixed as $\eta=\bar{\eta}\Delta t/(\Delta x)^2=0.08$ and $\sigma=\bar{\sigma}\Delta t/(\Delta x)^2=0.17$. This should not be interpreted as a rigorous prescription for fixing parameters but rather as a general rule to associate parameter values in a CDS model with those in the corresponding partial differential equation model. Finally, we discuss the most important parameter in (2), viz. α, which fixes the strength of the coupling between the order parameter and the velocity field. We have studied domain growth as modeled by (2) for a range of values of α. Stronger values of α accelerate the onset of the asymptotic time regime in which $L(t)\sim t$, whereas smaller values of α delay the onset of the asymptotic regime and give an extended period of slower growth. In the limit $\alpha\to 0$, we recover the Lifshitz-Slyozov growth law $L(t)\sim t^{1/3}$. In this paper, we present results for $\alpha=0.41$, which corresponds (in the prescription defined above) to $\bar{\alpha}=2$ in (1). Detailed results for different values of α will be published elsewhere [11].

We have implemented (2) with the above parameter values on an N^3 lattice with periodic boundary conditions. The results presented here are for the case $N=80$. We used a Siemens-Fujitsu VP100 vector processor and a single update of 80^3 lattice took approximately 1 CPU sec. All results presented here are for the case of a critical quench, where there are equal concentrations of both components in the mixture. Results for off-critical quenches will be presented later [11]. The time-dependent structure factor for the order parameter is defined as

$$S(\mathbf{k},t)=\langle\psi(\mathbf{k},t)\psi(-\mathbf{k},t)\rangle\,, \tag{3}$$

where $\psi(\mathbf{k},t)$ is the Fourier transform of the order parameter field at wave vector $\mathbf{k}$ and the angular brackets refer to an average over the ensemble of initial conditions. In the discrete case, we calculate (using the NAGLIB routine C06FJF) $\psi(\mathbf{k},t)$ as the discrete Fourier transform of the order parameter field $\psi(n,t)$. The wave vectors $\mathbf{k}$ lie in the first Brillouin zone of the lattice, viz. $\mathbf{k}=2\pi(n_x,n_y,n_z)/N$ where n_x, n_y, and n_z are integers between and including $-N/2$ and $N/2-1$. The structure factor is calculated as an average over 40 runs from different initial conditions, each of which consists of the order parameter and velocity field uniformly and randomly distributed about a zero background with amplitudes 0.05 and 0.1, respectively. The structure factor is normalized as $\sum_{\mathbf{k}}S(\mathbf{k},t)/N^3=1$. The vector function $S(\mathbf{k},t)$ is then spherically averaged to give the scalar function $S(k,t)$. The time-dependent characteristic length scale $L(t)$ is defined as the reciprocal of the first moment of the scalar structure factor $S(k,t)$, i.e., $L(t)=[\langle k\rangle(t)]^{-1}$, where

$$\langle k\rangle(t)=\frac{\sum_k kS(k,t)}{\sum_k S(k,t)}\,. \tag{4}$$

In (4), the sum over the k values is cut off at $k_{\max}=\pi\sqrt{3}/2$. We have confirmed that the structure factor has decayed sufficiently up to this wave vector so that

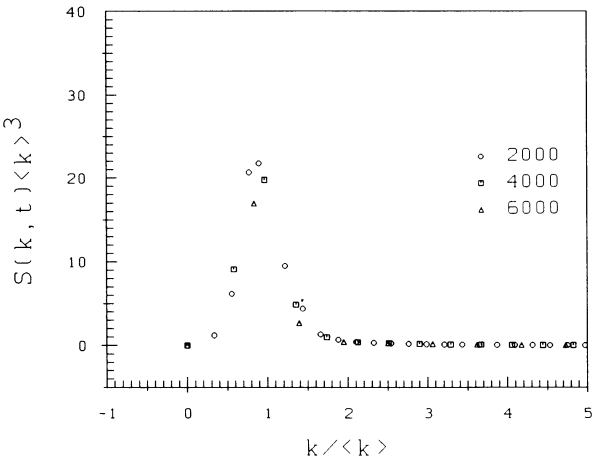

FIG. 1. Plot of $S(k,t)\langle k\rangle^3$ vs $k/\langle k\rangle$ for structure-factor data from a simulation of (2) on an 80^3 lattice with periodic boundary conditions. Data were obtained as averages over 40 independent runs from initial conditions corresponding to a homogeneous state. The structure factors are for update times 2000, 4000, and 6000 (marked by the symbols indicated).

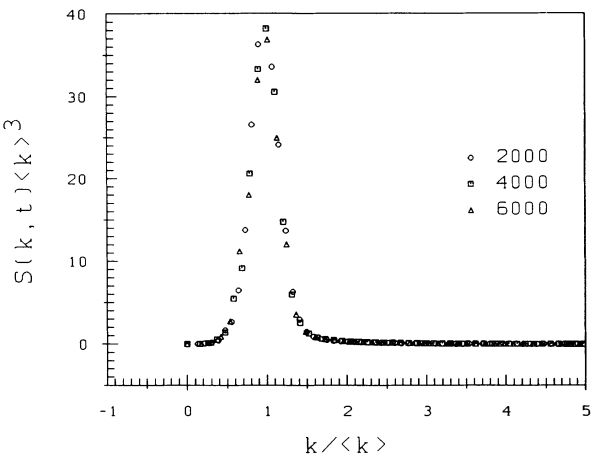

FIG. 2. Plot of $S(k,t)\langle k\rangle^3$ vs $k/\langle k\rangle$ for structure factor data from a CDS simulation of model B [viz. (2) with $\alpha=0$], implemented on an 80^3 lattice with periodic boundary conditions. The data were obtained by the same procedure as that described in the text for Fig. 1. The structure factors are for update times 2000, 4000, and 6000 (marked by the symbols indicated).

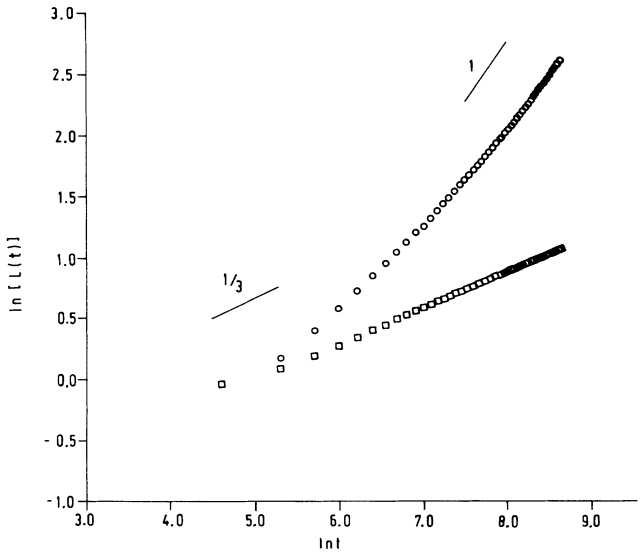

FIG. 3. Plot of the characteristic length scale $L(t)$ as a function of update time t for the case with hydrodynamics (marked by circles) and the case without hydrodynamics (marked by squares) on a double-logarithmic scale. The error bars on the data points are smaller than the symbol sizes. The solid lines have slopes of $\frac{1}{3}$ and 1, as marked.

the sum is a good approximation of the infinite integral.

For purposes of comparison, we have also performed a CDS simulation for the case without hydrodynamics (the so-called model B) on an 80^3 lattice. Our CDS model for the case without hydrodynamics is simply (2) with $\alpha=0$. We use the same values for A and D as in the case with hydrodynamics and the procedure whereby we obtained the structure factor and the characteristic length scale is the same as that just described.

Figure 1 shows the scaled structure factor $S(k,t)\times[\langle k\rangle(t)]^3$ plotted as a function of the scaled wave vector $k/\langle k\rangle(t)$ for data from update times 2000, 4000, and 6000. The excellent data collapse indicates that dynamical scaling [12] is valid and that the domain growth is characterized by a unique length scale. The form of the universal function in Fig. 1 is similar in shape to the case without hydrodynamics (shown in Fig. 2, for comparison) with the only major difference being that the peak of the scaled structure factor for model B is considerably higher. For earlier times, the form of the universal function for the case with hydrodynamics is identical to that for the case without hydrodynamics. Notice that there are no adjustable parameters in our definition of the structure factor.

Figure 3 shows the characteristic domain size $L(t)$ as a function of the update time t for our model (2) (marked by circles) and the case without hydrodynamics (marked by squares) on a double logarithmic scale. After an initial transient regime (which can be extended by using a smaller value of α), the domain size for the hydrodynamic case grows linearly in time, viz., $L(t)\sim t$. At times somewhat beyond those shown in Fig. 3, freezing sets in because of the finite size of the system. To ensure that our data are not affected by finite-size effects, we have also performed (less thoroughly) simulations on lattices of size 64^3 and 100^3. The results are the same as those presented here with the only difference being that the onset of freezing is delayed in the larger systems. Domain growth for the case without hydrodynamics is seen to obey the usual Lifshitz-Slozov growth law $L(t)\sim t^{1/3}$.

To summarize, we have reported the numerical observation of a temporally linear domain growth in a phenomenological model of segregating fluids. Our observation is facilitated by (i) the use of the simplest possible (minimal) model, and (ii) the use of a CDS model to accelerate the onset of the asymptotic regime. The onset of this asymptotically linear growth is determined by the strength of the coupling parameter α. For smaller values of α, we obtain a Lifshitz-Slyozov growth law for the early and intermediate times and this crosses over to the temporally linear growth reported here. We will provide detailed results in an extended publication, where we will present results for the decay of the current-current correlation function and also a comparison of our results with experiments on binary fluids.

Note added. After the submission of this manuscript, we have become aware of complementary works by Koga and Kawasaki [13] and Shinozaki and Oono [14]. Koga and Kawasaki [13] report a linear growth law in a binary fluid system, where the hydrodynamic interactions are described by the Oseen tensor. Shinozaki and Oono [14] study spinodal decomposition in a Hele-Shaw cell.

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