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Spinodal decomposition in a Hele-Shaw cell

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As an experimentally realizable two-dimensional fluid system, we perform a cell-dynamical system simulation of a binary incompressible fluid that undergoes spinodal decomposition after a critical quench in a Hele-Shaw cell. Fluidity enhances fluctuations of interfaces, resulting in a form factor that continuously depends on the fluidity of the system. In this system, finite size coupled with the effect of incompressible flow is found to have a severe accelerating effect on the growth law.

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The late-time behavior of systems undergoing spinodal decomposition is still an unresolved problem in nonequilibrium statistical physics [1]. Analytic theories of ordinary binary-alloy spinodal decomposition in the asymptotic, late-time regime are still unsatisfactory. Hydrodynamic interactions make this problem only more difficult. Two-dimensional (2D) fluid systems have been studied by various computational methods [2], but most of these systems appear to be either physically unrealistic or experimentally difficult to realize and none of these systems model incompressible flow. Recently, small-scale 3D fluid systems have also been studied [3], but we believe that careful large-scale studies of 2D fluid systems must be done to discover possible difficulties with future studies in 3D. As we show, we can expect very severe finite-size effects.

It is difficult to imagine a physical, free-standing 2D fluid system. A physically realizable situation may be a thin film of incompressible liquid sandwiched between two solid planes. Such a system is essentially a Hele-Shaw cell, used to study Saffman-Taylor fingering [4], for example, in which the fluid flow obeys Darcy's law. In this Rapid Communication, we present the results of a computational study modeling the spinodal decomposition of an incompressible symmetric binary fluid system after deep critical quench in a Hele-Shaw cell. Our main aim is to examine the effect of incompressible flow on the asymptotic behavior of spinodal decomposition. The numerical method is based on the cell-dynamical system (CDS) model of spinodal decomposition of a binary alloy [5]. This paper is a preparatory step to a large-scale 3D system with hydrodynamics.

If we allow a binary fluid mixture to undergo spinodal decomposition in a suitable Hele-Shaw cell, we may expect that the mean domain size will eventually be larger than that of the interplate spacing. At this point the fluid motion will be essentially 2D. Further the fluid motion should be slaved to the overall slow process of the phase separation. We expect in this situation that Darcy's law will be obeyed, with the modification that the force on the fluid will be due to the gradient in the chemical potential, $f=-\psi\nabla\delta\mathcal{F}_{\rm CG}/\delta\psi$ [6], where ψ, the order parameter, is proportional to the concentration difference between the two types of fluid, and where $\mathcal{F}_{\rm CG}$ is a coarse-grained phenomenological free energy. The velocity field will then obey

$$\mathbf{v}=-\frac{d^2}{\nu}\left[\nabla p+\psi\nabla\frac{\delta\mathcal{F}_{\rm CG}}{\delta\psi}\right],\quad \nabla\cdot\mathbf{v}=0\,, \tag{1}$$

where d is proportional to the interplate distance, ν is the viscosity, and p is the pressure determined by incompressibility. Since we study a thin fluid layer, the effect of differential wetting may be a serious experimental problem. This may be taken into account by a suitable choice of the free-energy functional and modeling of the wetting layer [7]. Here, we assume the effective free-energy functional is symmetric and that there is no preferential wetting of the cell walls.

Previous studies give us a workable cell-dynamical system for the spinodal decomposition of a binary-alloy system [5]. This model was successfully used to study the asymptotic form factor for a 3D binary-alloy model after critical quench [8]. The CDS model for spinodal decomposition of a binary-alloy system is

$$\psi_{t+1}(\mathbf{n})=\psi_t(\mathbf{n})+M[I_t(\mathbf{n})-\langle\langle I_t(\mathbf{n})\rangle\rangle]\,, \tag{2}$$

$$I_t(\mathbf{n})=D[\langle\langle\psi_t(\mathbf{n})\rangle\rangle-\psi_t(\mathbf{n})]+\mathcal{F}(\psi_t(\mathbf{n}))-\psi_t(\mathbf{n})\,. \tag{3}$$

The double angular brackets denote an average of a neighborhood of cells [9], D is a positive constant, and $\psi_t(\mathbf{n})$ denotes the order parameter in a cell at $\mathbf{n}$ at time t. The injective map $\mathcal{F}()$ models the local cell dynamics. $-I$ is an effective chemical potential. M is the mobility of the system. The map chosen for Eq. (3) is $\mathcal{F}(\psi)=A\tanh(B\psi)$, where $B=\tanh^{-1}(1/A)$. This map has stable fixed points at 1 and -1 for any $A>0$. The boundary conditions are periodic. We will call this model of spinodal decomposition the ordinary system.

To add fluid dynamics, we model the flow as described previously by Eq. (1). We will refer to the model as the Hele-Shaw system. The velocity of fluid flow in a cell is as follows:

$$\begin{aligned}\mathbf{v}_t(\mathbf{n})&=(1/-c^2)\{[\nabla]_d p_t(\mathbf{n})-\psi_t(\mathbf{n})[\nabla]_d I_t(\mathbf{n})\}\,,\\ [\nabla^2]_d p_t(\mathbf{n})&=[\nabla]_d\{\psi_t(\mathbf{n})[\nabla]_d I_t(\mathbf{n})\}\,,\end{aligned} \tag{4}$$

where $c^2 \propto (d^2/v)^{-1}$ is a positive constant. The $[\,]_d$ notation denotes the discrete version of the enclosed operator [10]. A standard fast-Fourier-transform technique solved the implicit pressure equation (4). We update the order-parameter field and then the velocity field is used to compute the next time step from the updated order-parameter field [11]:

$$\psi^*(\mathbf{n}) = \psi_t(\mathbf{n}) + M[I_t(\mathbf{n}) - \langle\langle I_t(\mathbf{n})\rangle\rangle] ,$$

$$\psi_{t+1}(\mathbf{n}) = \psi^* - [\nabla]_d[\psi^*(\mathbf{n})\mathbf{v}_t(\mathbf{n})] .$$

We study the form factor: $S(\mathbf{k},t) = \langle \psi_{\mathbf{k}}(t)\psi_{-\mathbf{k}}(t)\rangle$, where $\psi_{\mathbf{k}}$ is the Fourier transform of the order parameter and $\langle\,\rangle$ denotes ensemble averaging. Define $\hat{S}(k,t)$ as the circularly (in $\mathbf{k}$-space) averaged form factor after normalization by $\int d^3k\, S(\mathbf{k},t) = 1$. If we may assume that there is only one relevant length scale $l(t)$ [12], we can assert from dimensional analysis $\hat{S}(k,t) = l^d(t)F(x)$, where $F(x)$ is a dimensionless form factor, and $x = kl(t)$.

The most convenient characteristic length [13] can be defined in terms of

$$\langle k\rangle(t) = \frac{\int_0^\infty dk\, k\hat{S}(k,t)}{\int_0^\infty dk\, \hat{S}(k,t)} . \tag{5}$$

$\langle k\rangle(t)$ is essentially the reciprocal of $l(t)$, the characteristic length scale of the domains in a well decomposed system. If scaling holds, we may look for a growth exponent ϕ such that $\langle k\rangle(t) \sim t^{-\phi}$ in the limit of late times.

Using Eq. (1) we may follow Kawasaki and Ohta [14] and form an interfacial equation of motion. Dimensional analysis then gives $\phi = \frac{1}{3}$ as in the case of the ordinary spinodal decomposition for both bulk diffusion and the hydrodynamic flow. The relative effectiveness of the diffusive mechanism to the hydrodynamic mechanism is thus controlled by the factor Mc^2. Therefore, $F(x)$ may now be dependent on Mc^2.

We have studied several systems to investigate the finite-size effect and Mc^2 dependency. For parameters $A = 1.5$, $D = 0.4$, $M = 0.1$, $c^2 = 1.0$, $Mc^2 = 0.1$, we studied the following: two samples of sizes 128^2, one sample of size 256^2 and 512^2, and eight samples of size 384^2. Additionally we studied a system with parameters $A = 1.5$, $D = 0.4$, $M = 0.5$, $c^2 = 2.0$, $Mc^2 = 1.0$, eight samples of size 256^2. Experience has shown that the system self-averages well only for $k/\langle k\rangle > 2$ when examining the form factor and so one must have many samples to have an overall reliable form factor. However, $\ln[\langle k\rangle(t)]$ fluctuates mildly from sample to sample and usually one sample is sufficient to discern the gross behavior of $\ln[\langle k\rangle(t)]$. As a reference, we have also computed a 2D symmetric ordinary system with parameters $A = 1.3$, $D = 0.5$, $M = 1.0$, with 25 samples, of size 256^2. We will call the system with $Mc^2 = 0.1$ the high-fluidity system, and the one with $Mc^2 = 1.0$ the low-fluidity system.

Figure 1(a) exhibits the log-log behavior of $\langle k\rangle(t)$ for the high-fluidity ensemble of size 384^2. After a long transient phase, the slope appears to settle at $-\frac{1}{3}$ as expected from dimensional analysis. For the time range observed, systems of size 256^2 to 512^2 show almost identical behavior. However, there is a clear acceleration due to finite-size effects for the high-fluidity system of size 128^2. A mild acceleration was expected due to finite-size effects, however the actual effect is great.

We found a further hazard in simulating hydrodynamic effects. In Fig. 1(a) we also show behavior of $\langle k\rangle(t)$ of a system, $A = 1.5$, $D = 0.2$, $M = 0.1$, $c^2 = 1.0$, $Mc^2 = 0.1$, of

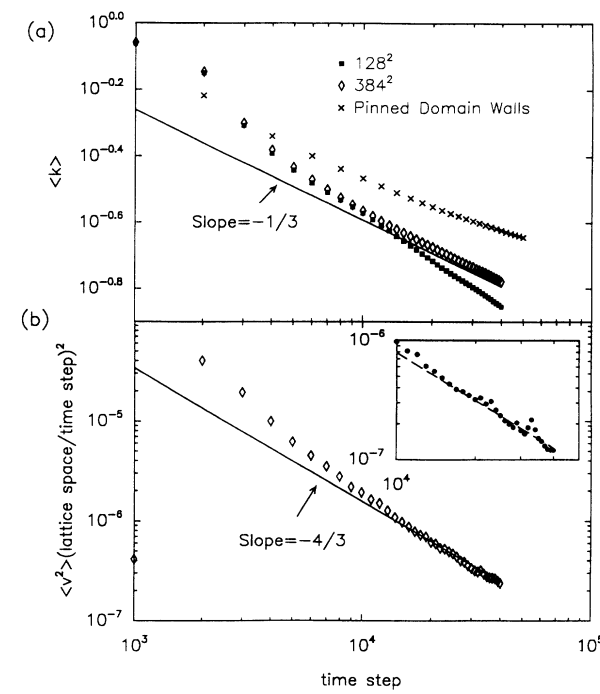

FIG. 1. (a) Log-log plot of $\langle k\rangle$ over time step. Solid squares and open diamonds are data from high-fluidity systems. The small system of size 128^2 (solid squares) shows a clear acceleration in growth law. The large system of size 384^2 (diamonds) obeys the expected $-\frac{1}{3}$ law. Samples of size 512^2 and 256^2 had nearly identical behavior as the 384^2 ensemble. The crosses exhibit behavior of a system that had numerically pinned domain walls. (b) Log-log plot of $\langle v^2\rangle$ over time step for high fluidity, size 384^2 system. Inset: The same plot for a single 384^2 sample over the time step 10000 to 40000.

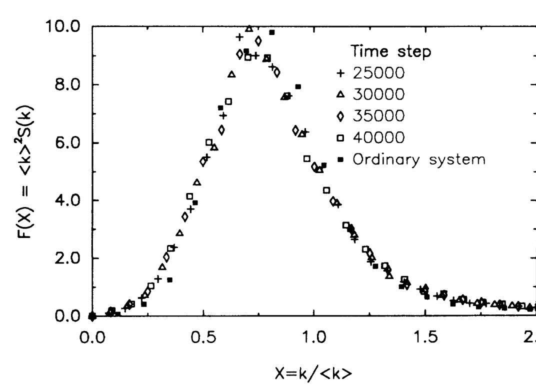

FIG. 2. Comparison of form factor for hardened data of high fluidity, size 384^2 systems over time and the hardened form factor for the late time (time step is 25000) ordinary system.

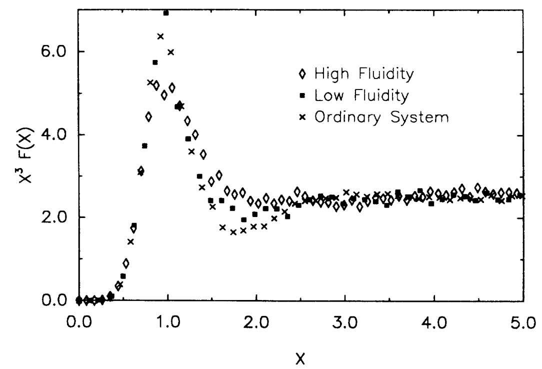

FIG. 3. Comparisons of plot of $x^3F(x)$, or Porod plot, using hardened data from high-fluidity system of size 384^2, low-fluidity system of size 256^2, and ordinary system.

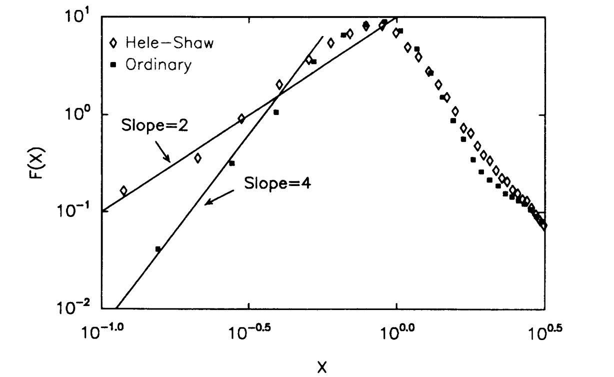

FIG. 4. Log-log plot of form factor for high-fluidity system of size 384^2 (time step is 40000) and the ordinary system (time step is 25000).

size 192^2, which exhibited a gradual pinning of the domain walls. This system gradually slowed down because low-curvature domain walls would become flat, freezing onto the underlying lattice. This type of effect has been seen in previous computational studies of coarse-grained models of late stage spinodal decomposition [15]. Essentially, this effect is due to "overcoarse graining" of the model. However, this effect was unexpected at the given parameters since the freezing or metastability did not appear in the ordinary model for these parameters. Flow appears to facilitate the system to rearrange itself locally, hence accentuating the tendency of domain walls to align with the computational lattice. To eliminate this effect, we broadened the walls slightly by increasing D, which is equivalent to decreasing the coarse-graining length.

The finite-size effect and numerical pinning have opposite effects on $\langle k\rangle(t)$. In fact, a very preliminary high-fluidity study on a small system nearly exhibited the expected $\langle k\rangle(t)\sim t^{-1/3}$, however this appears to have been to a fortuitous cancellation of effects. We expect similar problems in simulating 3D hydrodynamic systems.

If the $\mathbf{v}$ field scales asymptotically with the single length scale $l\propto\langle k\rangle^{-1}$, we expect from simple dimensional analysis that $\langle v^2\rangle\sim l^2/t^2\sim t^{-4/3}$, where the brackets denote sample average. Although we have obtained the scaled velocity-velocity correlation function $C_{vv}(\mathbf{r})\equiv\langle\mathbf{v}(\mathbf{r})\cdot\mathbf{v}(0)\rangle/\langle v^2\rangle$, here, we will only consider the time dependence of $\langle v^2\rangle$. We see in Fig. 1(b) that the $\langle v^2\rangle(t)$ log-log plot also reaches the expected scaling behavior at roughly the same time that $\langle k\rangle$ reaches scaling behavior. In the inset of Fig. 1(b), $\langle v^2\rangle$ of a single sample shows very intermittent behavior. The jumps appear to coincide with the rapid absorption of a droplet into a larger like-phase domain.

To numerically estimate the asymptotic form factor, we study the form factor of the order-parameter field after it is hardened by $\psi(x)\rightarrow\mathrm{sgn}[\psi(x)]$. This procedure has proved useful in a previous computational study of the 3D binary-alloy model [8]. Figure 2 shows that the hardened $S(k)$ scales well over a range of times, but also shows that the form factor is somewhat different from the ordinary case. Using a Porod plot (Fig. 3), we clarify the difference for $x\sim 2$. Note that the form factor has no discernible second hump in the high-fluidity case and that $F(x)$ continuously depends on the fluidity of the system.

For small wave vector, we examine the log-log plot of the unhardened form factor. Figure 4 shows that the relation $F(x)\sim x^4$, $x\ll 1$ [16] is not obeyed by the high-fluidity Hele-Shaw system. In fact, $F(x)\sim x^2$ seems more plausible. This could be a case of finite-size effects coupled to the wall fluctuations of large domains due to long-ranged hydrodynamic interaction.

Clearly there is a significant redistribution of structure to larger and smaller wave vector in the high-fluidity Hele-Shaw case from the ordinary case. By visual inspection, the patterns appear to be more "kneaded" and fragmented than that of the ordinary system. Small circular domains are trapped, and do not participate strongly with hydrodynamics. Their size is only changed by the slower (Lifshitz-Slyozov) diffusive mechanisms [17]. Hence there are many persistent small "drops" that probably enhance the form factor at small scales. Further, incompressible fluid dynamics allow small disturbances to have a large-scale effect. Visual inspection of patterns shows many events where an initially small change in pattern formation rapidly becomes a larger-scale reconfiguration of the domains about the small change, for example, the rapid merging of a droplet after contact with another domain. The higher the fluidity, the more pronounced should be these intermittent effects [18]. In 2D, the inevitable fragmentation of the domains, due to the constraints of dimension, allow opportunities for these events to exist in many places. For critical quenched systems in 3D, the domains may be always intertwined and percolated, but sufficiently off-critical systems may be affected.

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