Phase separation in two-dimensional fluids: The role of noise

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We use a lattice Boltzmann scheme within which the noise can be turned on and off to investigate the effect of stochastic terms on the phase ordering of a two-dimensional binary fluid. Sufficiently strong noise slows the growth in the hydrodynamic regime, changing the growth exponent $\alpha=2/3$ to $\alpha=1/2$. [S1063-651X(99)51105-7]

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I. INTRODUCTION

Binary mixtures of immiscible fluids, say A and B, quenched below the critical temperature phase separate into A- and B-rich domains that grow in time. Once integral domains have formed experimental and theoretical evidence shows that the typical domain size $R(t)$ grows as a power law with time t as [1]

$$R(t)\sim t^{\alpha}. \tag{1}$$

The exponent α is believed to be universal, depending only on the growth mechanism and not on the details of the particular system. The aim of this paper is to clarify the effect of noise on the value of α for phase separation in two-dimensional binary fluids.

The dynamics of binary fluids can be described on continuum length and time scales by the continuity, Navier-Stokes, and convection-diffusion equations [2],

$$\partial_t n+\partial_\alpha(nu_\alpha)=0, \tag{2}$$

$$\partial_t(nu_\beta)+\partial_\alpha(nu_\alpha u_\beta)=-\partial_\alpha P_{\alpha\beta}+\nu\nabla^2(nu_\beta)+\partial_\beta\{\lambda(n)\partial_\alpha(nu_\alpha)\}+\zeta_\beta, \tag{3}$$

$$\partial_t\varphi+\partial_\alpha(\varphi u_\alpha)=\Gamma\theta\nabla^2\Delta\mu-\theta\partial_\alpha\left(\frac{\varphi}{n}\partial_\beta P_{\alpha\beta}\right)+\xi, \tag{4}$$

where n is the total density of the fluid, u is the bulk fluid velocity, φ is the density difference between the two components, ν and λ are viscosities, and $\Gamma\theta$ is a diffusion coefficient. $P_{\alpha\beta}$ is the pressure tensor and $\Delta\mu$ the chemical potential difference between the A and B components. The noise terms ξ and $\vec{\zeta}$ are Gaussian distributed with zero average and correlations with strength representing temperature effects [2]. Greek indices are used to represent Cartesian directions and the usual repeated summation convention is assumed.

Several growth mechanisms are operative in binary fluids [1]. The first is Lifshitz-Slyozov growth, the relative diffusion of A and B atoms between domains [3]. This is described by Eq. (4) with $\vec{u}=0$, and simple dimensional analysis gives the correct growth exponent $\alpha=1/3$. The diffusive mechanism is dominant at early times and large viscosities when the Reynolds number is still too small to allow hydrodynamic flow to be important.

A second, faster growth process is curvature-driven hydrodynamic flow [4]. This corresponds to an exponent $\alpha=2/3$, which follows from dimensional analysis of the term on the left-hand side of the Navier-Stokes equation (3) and the pressure term on the right-hand side. This mechanism is important in systems with a low viscosity when the velocities are sufficiently developed. It leads to circular domains as flow is induced by the pressure difference between points of different curvature. However, it cannot decrease the number of domains in the system. Hence, it is important on increasingly long length scales: the circular domains remaining at shorter length scales grow more slowly by the Lifshitz-Slyozov mechanism [5]. This is in accord with a recent paper by Grant and Elder [6], who pointed out that the asymptotic growth exponent must be $\alpha\leqslant 1/2$.

In this Rapid Communication we shall be concerned with a third mechanism for growth in a two-dimensional binary fluid, which can result when stochastic terms are present in the Navier-Stokes equations. As first pointed out by San Miguel *et al.* [7] sufficiently large noise leads to a growth exponent $\alpha=1/2$. However, there is some confusion in the literature about whether and when noise changes the value of the critical exponent and about the mechanism for noise-driven growth.

The value $\alpha=1/2$ has been observed in computer simulations of domain growth in binary fluids using molecular dynamics [8]. $\alpha=2/3$ was seen in lattice Boltzmann simulations of growth where noise was absent [9]. However, the situation is not entirely clear. Lookman *et al.* [10] performed simulations without noise that gave $\alpha=1/2$ and calculations using lattice gas cellular automata where noise is an integral part of the simulation gave both $\alpha=1/2$ [11] and $\alpha=2/3$ [12].

To investigate these points we consider domain growth in a two-dimensional binary fluid using lattice Boltzmann simulations [13], which have the advantage that it is possible to include stochastic terms that can be switched on and off [14]. In this way we are able to test directly the effect of noise on the domain growth. We consider both noise that couples to the pressure and noise that couples to the chemical potential.

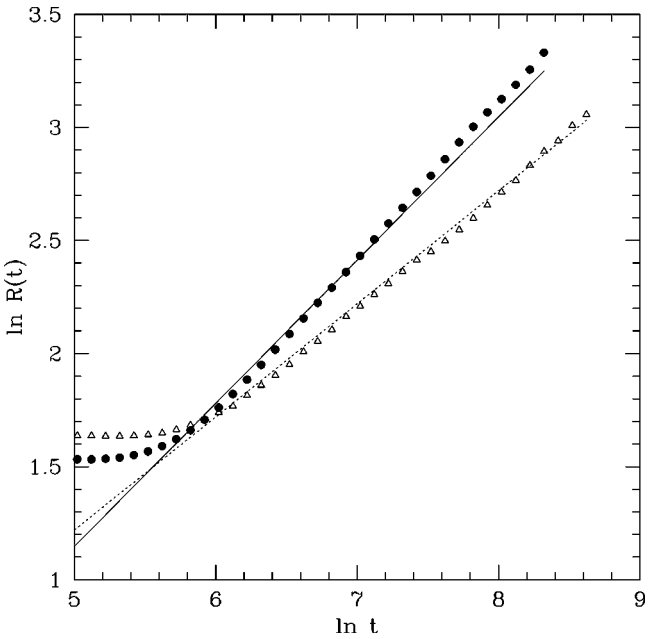

FIG. 1. Growth of typical domain size R with time t for a two-dimensional binary fluid for ●, no noise ($\tau_1=0.8$); △, a stochastic term in the pressure tensor [$\tau_1=0.8$; var(ζ)$=0.005$]. The lines correspond to growth exponents $\alpha=\frac{2}{3}$ (solid line) and $\alpha=\frac{1}{2}$ (dashed line).

We first summarize the lattice Boltzmann approach and describe how stochastic terms are introduced into the simulations. We then present the results of the computer simulations and discuss the mechanism by which noise changes the value of the growth exponent.

II. MODEL

We use a nine-velocity lattice Boltzmann model described in detail in Ref. [15]. The lattice Boltzmann method is essentially a finite difference scheme for solving the continuum equations (2)–(4) [13]. One-particle, directional distribution functions $f_i(\vec{x},t)$ and $g_i(\vec{x},t)$ are defined at each site $\vec{x}$ of a square lattice for each of nine directions i corresponding to lattice vectors $\vec{e}_i$. These are related to the physical variables by

$$n=\sum_i f_i, \quad \varphi=\sum_i g_i, \quad nu_\alpha=\sum_i f_i e_{i\alpha}. \tag{5}$$

The $\{f_i\}$ and $\{g_i\}$ evolve by streaming along direction i followed by a collision step that conserves density, momentum, and density difference. The evolution equations are

$$f_i(\vec{x}+\vec{e}_i\Delta t,t+\Delta t)-f_i(\vec{x},t)=-\frac{1}{\tau_1}(f_i-f_i^0+\zeta_i), \tag{6}$$

$$g_i(\vec{x}+\vec{e}_i\Delta t,t+\Delta t)-g_i(\vec{x},t)=-\frac{1}{\tau_2}(g_i-g_i^0+\xi_i), \tag{7}$$

identical to those of Ref. [15] except for the addition of the stochastic terms ζ_i and ξ_i [14]. The right-hand sides of Eqs. (6) and (7) are linearized collision operators with the source terms f_i^0 and g_i^0 chosen first so that the conservation laws are obeyed and second so that in thermodynamic equilibrium the model fluid minimizes a chosen input free energy density [16]. In these simulations we use

$$\Psi=\int d\vec{r}\left\{\frac{a}{2}(\varphi)^2+\frac{b}{4}(\varphi)^4+\frac{\kappa}{2}(\nabla\varphi)^2\right\}, \tag{8}$$

with $a=-1, b=1$, and $\kappa=1$. The pressure tensor $P_{\alpha\beta}$ and chemical potential $\Delta\mu$ which are calculated [17] from Ψ appear in the Navier Stokes equation (3) and convection-diffusion equation (4), respectively.

The lattice Boltzmann scheme reproduces to second order the continuum equations (2)–(4) with viscosities and diffusivity related to the relaxation parameters τ_1 and τ_2 through

$$\nu=\frac{(2\tau_1-1)}{6}(\Delta t)c^2, \quad \lambda(n)=\left(\tau_1-\frac{1}{2}\right)\Delta t\left(\frac{c^2}{2}-\frac{dp_0}{dn}\right),$$

$$\theta=(\Delta t)c^2(\tau_2-1/2). \tag{9}$$

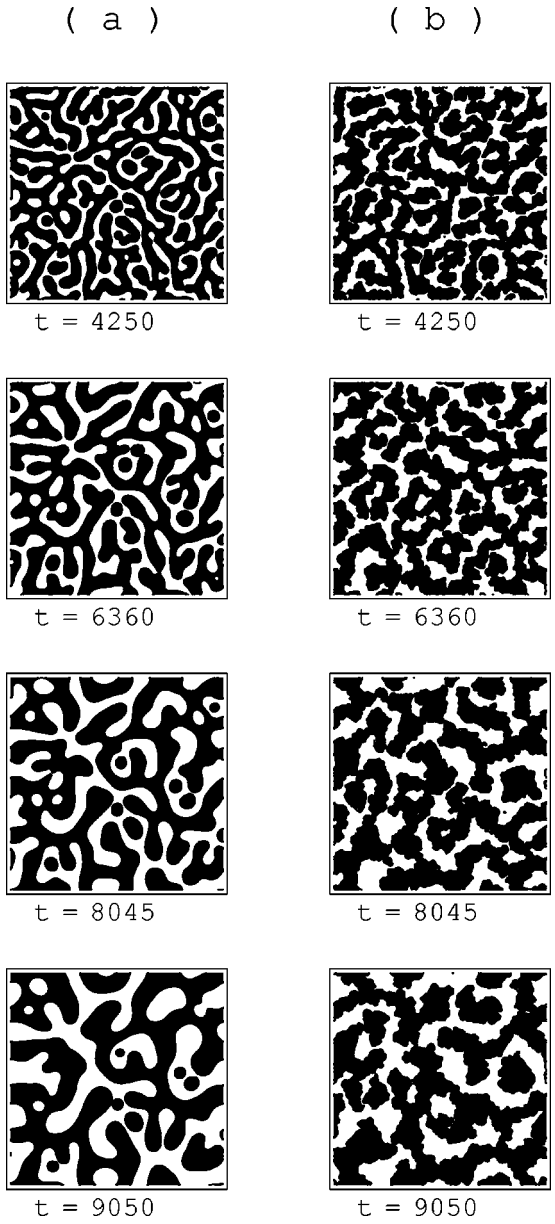

FIG. 2. Snapshots of the domain growth of a binary fluid comparing the evolution for (a) no noise, and (b) a stochastic term in the pressure tensor [$\tau_1=0.8$; var(ζ)$=0.005$ in (b)].

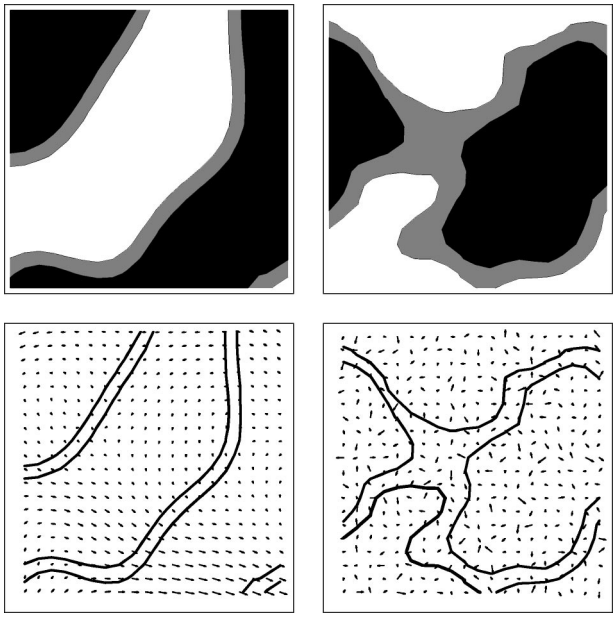

FIG. 3. Local features of the growing domain pattern and the corresponding velocity field for no noise (left-hand-column), and a stochastic term in the pressure tensor (right-hand-column). The velocity field is randomized by the noise [$\tau_1=0.8$, var(ζ)$=0.005$ in (b)].

We use $c=1$, $\triangle t=0.01$, $\tau_2=(1+1/\sqrt{3})/2\simeq 0.79$, and $\Gamma=1$. Different values of τ_1 are used in different runs to control the value of the viscosity.

We now describe how we choose the noise term ζ_i in Eq. (6). σ_{xx} and σ_{xy} are chosen as independent Gaussian variables with a given variance. We take $\sigma_{yy}=-\sigma_{xx}, \sigma_{xy}=\sigma_{yx}$ and put

$$\zeta_0=0, \quad \zeta_1=\zeta_3=\frac{1}{2}\sigma_{xx}, \quad \zeta_2=\zeta_4=\frac{1}{2}\sigma_{yy}, \tag{10}$$

$$\zeta_5=-\zeta_6=\zeta_7=-\zeta_8=\frac{1}{4}\sigma_{xy}. \tag{11}$$

It can be easily verified that for the nine-velocity model

$$\sum_i \zeta_i=0, \quad \sum_i \zeta_i e_{i\alpha}=0, \tag{12}$$

and hence that the conservation of mass and momentum is unaffected by the noise. The contribution of the noise to the pressure tensor is

$$P_{\alpha\beta}\equiv\sum_i \zeta_i e_{i\alpha}e_{i\beta}=\sigma_{\alpha\beta}. \tag{13}$$

In particular, ζ_i with $1\leqslant i\leqslant 4$ contributes to P_{xx} and P_{yy}, while ζ_i with $5\leqslant i\leqslant 8$ contributes to P_{xy}. The ξ are defined similarly. This allows us to tune the strength of the fluctuations, which is measured by the variance of the random variables, var(ζ).

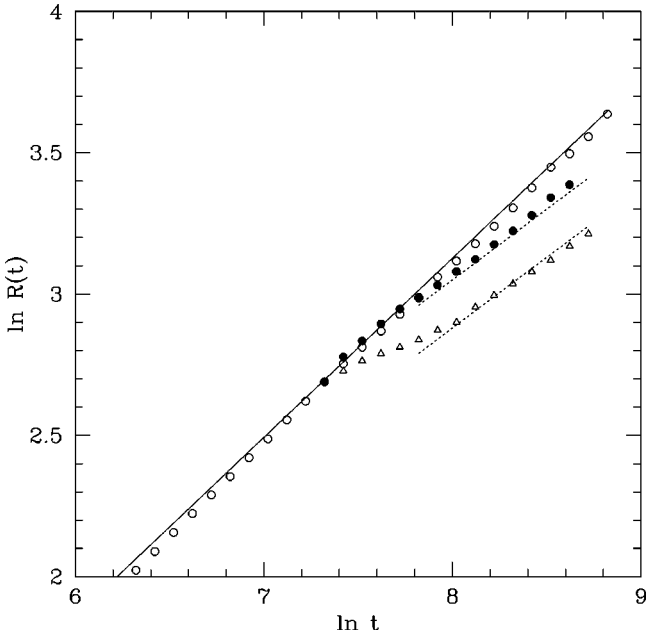

FIG. 4. Growth of typical domain size R with time t for a two-dimensional binary fluid when a stochastic term coupling to the pressure tensor is switched on during the growth process. $\tau_1=0.8$. ○, no noise; ● var(ζ)$=0.003$; △, var(ζ)$=0.005$. The lines correspond to growth exponents $\alpha=\frac{2}{3}$ (solid line), and $\alpha=\frac{1}{2}$ (dashed line).

III. RESULTS

In all of the simulations reported the system was initialized with $n=1.0$ and φ chosen randomly between -0.5 and 0.5 and then quenched to a final state defined by parameters $a=-1, b=\kappa=1$. The density n remains an essential constant throughout the fluid in all of the cases considered. The system size was 256×256 and simulations were typically run for 10^5 time steps. Consider first $\xi=0, \zeta\neq 0$; that is, the addition of a noise term in the pressure tensor which directly affects the velocity field. Figure 1 shows the average size of domains following a quench measured by the inverse of the first moment of the structure factor for two sets of runs. All of the parameters were the same except for the noise. Each set consists of three different runs, and the results of each set are averaged in the figure. For no noise the usual growth exponent $\alpha=2/3$ is clearly seen. In the simulations with noise the growth law is $\alpha=1/2$.

Pictorial snapshots of the time evolution of runs with and without noise are compared in Fig. 2. The immediate conclusion is that noise increases the roughness of the surfaces of the domains. This corresponds, on a mesoscopic scale, to the randomization of the velocity field, as shown in Fig. 3.

The crossover in the value of the growth exponent from 2/3 to 1/2 occurs because the noise destroys the driving force for growth, which results from the pressure difference between points of different curvature on a domain surface. The velocity field is now driven by the noise rather than the Laplace pressure. Hence, the exponent $\alpha=1/2$ can be deduced from dimensional analysis balancing the noise term $\sim R^{-1}(R^2t)^{-1/2}$ and the inertial terms on the left-hand side of Eq. 3 $\sim Rt^{-2}$.

The crossover between $\alpha = 2/3$ and 1/2 is expected to occur when

$$\Delta P \sim \sigma/R \sim \mathrm{var}(\zeta), \tag{14}$$

where ΔP is the Laplace pressure and σ is the surface tension. This is illustrated in Fig. 4. A simulation was run without noise until the 2/3 growth law was well established. The noise was then turned on and further evolution of the run was compared with var$(\zeta) = 0$, 0.003, and 0.005. With no noise the $\alpha = 2/3$ growth persisted as expected. For var$(\zeta) = 0.005$ the exponent rapidly crossed over to $\alpha = 1/2$. For var$(\zeta) = 0.003$ the crossover was slower. The approximate values of σ and R at the crossover are $R \sim 16, \sigma \simeq 0.05$ for var$(\zeta) = 0.005$ and $R \sim 18$, $\sigma \simeq 0.04$ for $(\zeta) = 0.003$, consistent with Eq. (14).

To check the role of the noise in the domain growth we turned off the Laplace pressure artificially by removing the derivative terms in the pressure tensor $P_{\alpha\beta}$. The system grew very slowly, approaching an exponent consistent with 1/3. Therefore, the value 1/2 found in [14] remains puzzling.

Finally, we consider the effect of the noise term ξ, which couples to the chemical potential and hence directly affects the order parameter φ rather than the velocity field. Figure 5 shows the evolution of the domain size for three different sets of parameter values starting from the same initial conditions. These correspond to (i) low viscosity ($\tau_1 = 0.8$), low noise [var$(\xi) = 0.001$]; (ii) high viscosity ($\tau_1 = 50$), high noise [var$(\xi) = 0.01$]; (iii) low viscosity ($\tau_1 = 0.8$), high noise [var$(\xi) = 0.01$]. The main effect of the noise is at early time when domains are forming. In the noisier systems the order parameter takes longer to reach its equilibrium value within each domain.

Once the domains have formed the noise has no observable effect on the evolution. In the low viscosity simulations inertial flow gives an exponent 2/3 independent of the strength of the noise. This is because noise in the chemical potential difference is local and does not destroy the flow field. Similarly, for the high viscosity simulation where diffusive growth is expected, $\alpha = 1/3$. This is also as expected: noise is known to be irrelevant for Lifshitz-Slyozov growth [1].

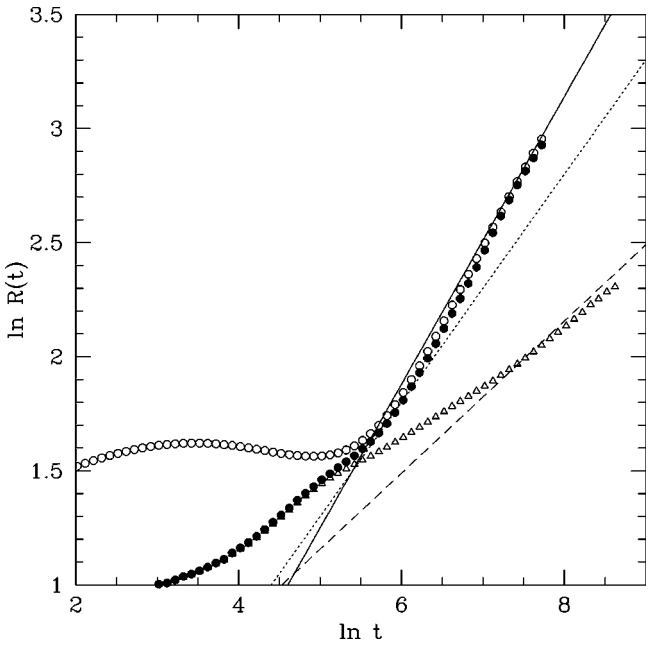

FIG. 5. Growth of typical domain size with time for a binary fluid with a stochastic term in the chemical potential difference: ○, low noise [var$(\zeta) = 0.001$], low viscosity ($\tau_1 = 0.8$); ●, high noise [var$(\zeta) = 0.01$], low viscosity ($\tau_1 = 0.8$); △, high noise [var$(\zeta) = 0.01$], high viscosity ($\tau_1 = 50$). The noise inhibits the initial formation of the domains, but neither the diffusive growth law $\alpha = 1/3$ expected at high viscosities nor the hydrodynamic growth with $\alpha = 2/3$ expected at low viscosities is altered. The straight lines correspond to $\alpha = 2/3$ (solid), $\alpha = 1/2$ (short dashed), and $\alpha = 1/3$ (long dashed).

We conclude that noise can destroy curvature-driven hydrodynamic growth and hence the corresponding $\alpha = 2/3$ growth regime. The system crosses over to a regime with $\alpha = 1/2$, where the velocity field is driven by the noise.

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[1] A.J. Bray, Adv. Phys. **43**, 357 (1994).
[2] B.I. Halperin and P.C. Hohenberg, Rev. Mod. Phys. **49**, 435 (1977).
[3] I.M. Lifshitz and V.V. Slyozov, J. Phys. Chem. Solids **19**, 35 (1961).
[4] H. Furukawa, Physica A **204**, 237 (1994).
[5] A.J. Wagner and J.M. Yeomans, Phys. Rev. Lett. **80**, 1429 (1998).
[6] M. Grant and K.R. Elder, Phys. Rev. Lett. **82**, 14 (1999).
[7] M. San Miguel, M. Grant, and J.D. Gunton, Phys. Rev. A **31**, 1001 (1985).
[8] E. Velasco and S. Toxvaerd, Phys. Rev. E **54**, 605 (1996).
[9] W.R. Osborn, E. Orlandini, M.R. Swift, J.M. Yeomans, and J.R. Banavar, Phys. Rev. Lett. **75**, 4031 (1995).
[10] T. Lookman, Y. Wu, F.J. Alexander, and S. Chen, Phys. Rev. E **53**, 5513 (1996).
[11] A.N. Emerton, P.V. Coveney, and B.M. Boghosian, Phys. Rev. E **55**, 708 (1997).
[12] S. Bastea and J.L. Lebowitz, Phys. Rev. E **52**, 3821 (1995).
[13] S. Chen and G.D. Doolen, Annu. Rev. Fluid Mech. **30**, 329 (1998).
[14] A.J.C. Ladd, Phys. Rev. Lett. **70**, 1339 (1993); **271**, 285 (1994).
[15] G. Gonnella, E. Orlandini, and J.M. Yeomans Phys. Rev. E **58**, 480 (1998).
[16] M.R. Swift, E. Orlandini, W.R. Osborn, and J.M. Yeomans, Phys. Rev. E **54**, 5041 (1996).
[17] A.J.M. Yang, P.D. Fleming, and J.H. Gibbs, J. Chem. Phys. **64**, 3732 (1976).