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Phase separation in two-dimensional binary fluids

M. San Miguel,* Martin Grant, and J. D. Gunton
Physics Department, Temple University, Philadelphia, Pennsylvania 19122
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Various domain-growth mechanisms in phase-separating binary fluids are discussed. A tube hydrodynamic instability for concentrated mixtures is studied through a linear stability analysis. A quantitative discussion of the three-dimensional linear growth law is given. For two-dimensional concentrated mixtures we argue that domains grow in time with a diffusive growth law $t^{1/2}$. No crossover to a linear growth law is expected, in contrast to $d=3$. This is in apparent agreement with a molecular-dynamics simulation of a $d=2$ pure fluid. For two-dimensional dilute mixtures we argue that a $t^{1/2}$ diffusive behavior crosses over to a $t^{1/3}$ Lifshitz-Slyozov growth in the latter stages of phase separation.

I. INTRODUCTION

The dynamics of a system undergoing a first-order phase transition (spinodal decomposition or nucleation and growth) is a complicated nonlinear problem.[1,2] Different and competing physical mechanisms drive the system to its final equilibrium state. Progress has been made by focusing on particular processes which can dominate the dynamics over a given time interval.

Our main interest here is phase-separating binary fluids.[3,4] In an analysis of three-dimensional binary fluids, Siggia[5] has identified several mechanisms for domain growth, in the latter stages of phase separation. (See also recent discussions by Kawasaki and Ohta.[6,7]) The scenario he proposed for three-dimensional fluids is as follows. If the fluid mixture is far from critical concentration (that is, if one species dominates), after formation of domains of size R, there is an initial growth by diffusive droplet coalescence. This mechanism leads to a growth law $R \sim t^{1/3}$, where t is time. Following this, the evolution would be dominated by an evaporation-condensation process, given by the classic analysis of Lifshitz and Slyozov.[8,9] This also gives a $t^{1/3}$ behavior, although the amplitude differs from that of coalescence. On the other hand, for concentrated mixtures, spinodal decomposition results in the initial formation of an interconnected structure. Growth proceeds initially by interface diffusion resulting in a $t^{1/3}$ behavior. Siggia predicted that a hydrodynamic flow effect would dominate following the diffusive behavior, giving rise to a growth law $R \sim t$ corresponding to a breakup of the interconnected structure. Finally, there is a crossover to gravitational coalescence in the very late stages. This hydrodynamic effect, which we will study in more detail below, involves the "necking down" of the tubularlike interconnected structure, which forms during spinodal decomposition, through the viscous response of the velocity field to the capillary pressure at an interface. We note that light scattering experiments on binary fluids[3,4] are in reasonable agreement with the general picture outlined above.

In this paper, motivated by the possibility of doing experiments on two-dimensional fluids,[10] we reexamine Siggia's scenario for dimension $d=2$. (Note, though, that experiments would probably consider monolayers suspended on fluids. Such monolayers could have different dynamical properties than the two-dimensional fluid mixture considered here.) We present an alternative treatment of Siggia's argument for the tube instability in $d=3$, in Sec. II, and discuss related work in the hydrodynamics literature due to Rayleigh[11] and Tomotika.[12] In particular, we argue that a nonlinear growth law can be estimated from linear stability analysis, as explained in the text. In Sec. III we study the analog of a tube in $d=2$. We find that "strips" are stable under small perturbations, in contrast to the three-dimensional result. Finally, in Sec. IV we enumerate possible growth mechanisms for two-dimensional fluids. For concentrated mixtures, we suggest that the phase separation proceeds via a $t^{1/2}$ interface diffusion mechanism followed by a $t^{1/2}$ droplet-coalescence growth. On the basis of the linear stability analysis of Sec. III, no crossover to a linear growth law is expected. For dilute mixtures we expect behavior similar to that given previously for $d=3$, except that, in $d=2$, the diffusive droplet coalescence would be a $t^{1/2}$ behavior. We also briefly discuss phase separation in a $d=2$ pure fluid.[13,14] The predicted absence of a linear growth law is in apparent agreement with a recent molecular-dynamics simulation of a $d=2$ liquid-vapor system.[14] Our analyses of binary and pure fluids, however, do not rule out the possibility of other growth mechanisms. Certainly, therefore, an experimental study would be of great interest.

II. TUBE INSTABILITY IN THREE DIMENSIONS

Before presenting the analysis for two dimensions, we will briefly discuss the $d=3$ results. We attempt to clarify the mechanism for the breakup of the interconnected structure originally proposed by Siggia. The hydrodynamic equations for an incompressible fluid of velocity $\mathbf{v}$ and density ρ are[15]

$$\partial_t \mathbf{v}(\mathbf{r},t)+(\mathbf{v}\cdot\nabla)\mathbf{v}=-\frac{1}{\rho}\nabla P(\mathbf{r},t)+\frac{\eta}{\rho}\nabla^2\mathbf{v}(\mathbf{r},t) , \quad (2.1)$$

which is the Navier-Stokes equation, where P is the pressure and η is the bulk viscosity, and

$$\nabla\cdot\mathbf{v}=0\ , \tag{2.2}$$

which is the continuity equation. We will assume that concentration gradients enter only through the "boundary conditions"[16–18] at the interfaces between the two fluids. That boundary condition is given by the equation of state for the surface, i.e., the capillary pressure (in $d=3$) (Ref. 15)

$$P_\sigma=\sigma\left[\frac{1}{R_1}+\frac{1}{R_2}\right] \tag{2.3}$$

called the Laplace-Young equation, where σ is the surface tension, and R_1 and R_2 are the principle radii of curvature at a given point on an interface.

Siggia's[5] arguments can be paraphrased as follows (see also the earlier work of Levich,[15] and Cahn and Moldover[19]). Let us idealize the interconnected structure as a long fluid tube of radius R. From Eq. (2.3) the capillary pressure will be roughly σ/R. However, this must match the pressure in the bulk fluid given by the Navier-Stokes equation [Eq. (2.1)], which is roughly $\eta\nabla\cdot\mathbf{v}$, if the viscous term dominates. Dimensional analysis then leads to the growth law

$$R\sim\frac{\sigma}{\eta}t\ . \tag{2.4}$$

A quantitative understanding of the origin of this hydrodynamic flow can be obtained by linear stability analysis. Indeed, Rayleigh[11] and Tomotika[12] studied the stability of a long cylindrical tube of fluid against long-wavelength fluctuations. These treatments solve the linearized hydrodynamic equations, obtained from Eqs. (2.1)—(2.3) above, for that geometry (see Fig. 1). (We extend Tomotika's analysis to $d=2$ in Sec. III.) Tomotika found that a tube of radius a was unstable for wave numbers $k<1/a$. The analysis gives exponential growth $e^{\omega t}$, where

$$\omega=\frac{\sigma}{2\eta a}(1-k^2a^2)\Phi_3(\eta/\eta',ka)\ , \tag{2.5}$$

Φ_3 is a positive definite quantity, and η' is the viscosity of the fluid surrounding the tube. The most unstable mode, that where $(1-k^2a^2)\Phi_3(\eta/\eta',ka)$ is maximized for a given η/η', is given by

$$k_{\max}a=\alpha\ , \tag{2.6}$$

where α is a function of η/η' alone. For $\eta/\eta'\simeq1$, one finds that $\alpha\simeq0.6$. An early application of these ideas to phase separation in polymer-polymer blends is due to McMaster.[20] It is worth noting that the instability disappears in the limit $a\rightarrow\infty$. This is because the instability explicitly involves the interaction of the cylindrical surface with itself. Thus to describe this growth mechanism, one must incorporate the correct geometry into an interfacial dynamical approach.[6,7]

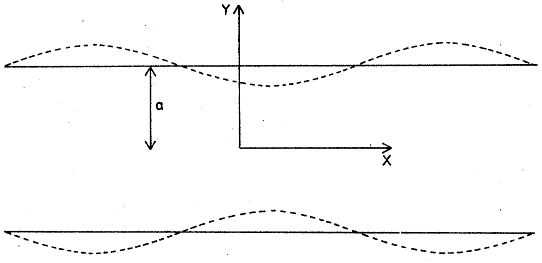

FIG. 1. Geometry for linear stability analysis in Sec. III. Fluid of density ρ is in the region $|y|<a$, surrounded by fluid of density ρ'. Broken lines schematically indicate a small perturbation. (For $d=3$ this figure corresponds to a cross section of a long cylinder.)

We will now argue that a growth law can be obtained from the linear stability analysis. This is related to a procedure used earlier by Langer.[21,22] Thus we are explicitly assuming that all the relevant physics is present, albeit in an approximate form, in the linear treatment. The outcome of linearizing a dynamical equation around some unstable value R_0 is an exponential growth characterized by a time constant τ, i.e.,

$$\frac{d\,\delta R(t)}{dt}=\frac{1}{\tau}\delta R(t)\ , \tag{2.7}$$

where $\delta R(t)\equiv R(t)-R_0$. This time constant is often of the form

$$\tau(R_0)=R_0^n/\lambda\ , \tag{2.8}$$

where λ is some constant, independent of R_0. Consider Eq. (2.7) at $t=0$. If $R(t=0)$ is proportional to R_0, then

$$\left.\frac{dR}{dt}\right|_{t=0}\sim\frac{\lambda}{R^{n-1}(t=0)}\ . \tag{2.9}$$

The proportionality constant is difficult to estimate since it would correspond to fluctuations in the initial nonequilibrium state, for our particular problem. Assuming that Eq. (2.9) can be extrapolated to finite times, which is a nontrivial assumption [or equivalently, that τ is the characteristic time scale for evolution in Eq. (2.8)], we have

$$R^n(t)\sim\lambda t\ . \tag{2.10}$$

This procedure gives sensible results for other related problems.[23]

Thus, considering the most unstable mode in Eq. (2.5), we obtain

$$R(t)\sim\frac{\sigma}{2\eta}(1-\alpha^2)\Phi_3(\eta/\eta',\alpha)t \tag{2.11}$$

for the tube instability. Pure dimensional analysis does not give the dependence upon the dimensionless ratio η/η'. For $\eta/\eta'\simeq1$, say, one obtains $R\simeq0.04(\sigma/\eta)$.[24] Siggia's estimate for the amplitude was 0.1, while the experimental result of Wong and Knobler[4] was 0.0013 ± 7. The crudeness of our treatment precludes an explanation of the discrepancy between theory and experiment.

III. LINEAR STABILITY ANALYSIS IN TWO DIMENSIONS

We will now extend Tomotika's analysis to a two-dimensional strip. The geometry is shown in Fig. 1. As stated earlier, our analysis is motivated by the possibility of doing experiments on two-dimensional binary fluids.[10] We will find below that the strip geometry is stable under small perturbations. Thus the $R \sim t$ mechanism may be inoperative in two-dimensional phase separation. This is, perhaps, not too surprising if one recalls that the analog of a two-dimensional strip in $d=3$ can be a thin slab, as well as a cylindrical tube.

Consider Eqs. (2.1) and (2.2). We will neglect the convective nonlinearity in the Navier-Stokes equation, although these effects could cause a subtle renormalization of the viscosity.[25] Then both the strip in the region $|y|<a$, characterized by a density ρ and kinematic viscosity $\nu=\eta/\rho$, and the surrounding fluid (of density ρ' and kinematic viscosity ν') satisfy linear equations.

The continuity equation (2.2) is satisfied by introducing a potential $\psi(x,y)$ such that

$$v_x=\frac{\partial\psi}{\partial y} \quad \text{and} \quad v_y=-\frac{\partial\psi}{\partial x}\ , \tag{3.1}$$

where $\mathbf{v}=(v_x,v_y)$. Eliminating the pressure with (3.1), Eq. (2.1) becomes[15]

$$(\partial_t-\nu\nabla^2)\nabla^2\psi(x,y,t)=0\ . \tag{3.2}$$

We look for a solution of (3.2) of the form

$$\psi(x,y,t)=\psi_1(x,y,t)+\psi_2(x,y,t)\ , \tag{3.3}$$

where

$$\nabla^2\psi_1=0\ , \tag{3.4}$$

$$(\partial_t-\nu\nabla^2)\psi_2=0\ . \tag{3.5}$$

A normal-mode analysis of (3.3)—(3.5) leads to

$$\psi_1(x,y,t)=A_1e^{ky+\omega t+ikx}+B_1e^{-ky+\omega t+ikx}\ , \qquad y>a \tag{3.6}$$

$$\psi_2(x,y,t)=A_2e^{K_1'y+\omega t+ikx}+B_2e^{-K_1'y+\omega t+ikx}\ , \qquad y>a \tag{3.7}$$

where

$$(K_1')^2=k^2+\frac{\omega}{\nu'}\ . \tag{3.8}$$

The same solution is valid for $y<-a$ with A_1, A_2, B_1, and B_2 replaced, respectively, by $\overline{A}_1$, $\overline{A}_2$, $\overline{B}_1$, and $\overline{B}_2$. The solution for $|y|<a$ is given by (3.6) and (3.7) with A_1, A_2, B_1, and B_2 replaced, respectively, by A_1', A_2', B_1', and B_2' and $(K_1')^2$ replaced by

$$K_1^2=k^2+\frac{\omega}{\nu}\ . \tag{3.9}$$

We require a symmetry property for the general solution $\psi(x,y,t)=\psi(x,-y,t)$. This gives $\overline{A}_1=B_1$, $\overline{A}_2=B_2$, $\overline{B}_1=A_2$, $\overline{B}_2=A_2$, $B_1'=A_1'$, and $B_2'=A_2'$. We also require ψ to be finite at $y=\pm\infty$, which implies $A_1=A_2=0$. The four remaining independent constants B_1, B_2, B_1', and B_2' are determined by boundary conditions at $y=a$.

(i) Continuity of v_x at $y=a$ gives

$$2B_1'ka\sinh(ka)+2B_2'K_1a\sinh(K_1a)+B_1kae^{-ka}+B_2K_1'ae^{-K_1'a}=0\ . \tag{3.10}$$

(ii) Continuity of v_y at $y=a$ gives

$$2B_1'\cosh(ka)+2B_2'\cosh(K_1a)-B_1e^{-K_1'a}-B_2e^{-K_1'a}=0\ . \tag{3.11}$$

(iii) Continuity at $y=a$ of the tangential stress[15]

$$\sigma_{xy}=\eta\left[\frac{\partial v_x}{\partial y}+\frac{\partial v_y}{\partial x}\right] \tag{3.12}$$

gives

$$4B_1'\frac{\eta}{\eta'}k^2\cosh(ka)+2B_2'\frac{\eta}{\eta'}(k^2+K_1^2)\cosh(K_1a)-2B_1k^2e^{-ka}-B_2[(K_1')^2+k^2]e^{-K_1a}=0\ . \tag{3.13}$$

(iv) The difference in the normal stress σ_{yy} is due to capillary pressure P_σ,

$$\sigma_{yy}(y\to a+)-\sigma_{yy}(y\to a-)=P_\sigma\ , \tag{3.14}$$

where[15]

$$\sigma_{yy}=-P+2\eta\frac{\partial v_y}{\partial y}\ . \tag{3.15}$$

For the pressure P the normal-mode analysis implies

$$P=P_0e^{\omega t+ikx}\ . \tag{3.16}$$

Substituting in (2.1) with (3.1) and (3.3) gives

$$\frac{P}{\rho}=\frac{i\omega}{k}\frac{\partial\psi}{\partial y}-\frac{i\nu}{k}(K_1^2-k^2)\frac{\partial\psi_2}{\partial y}\ . \tag{3.17}$$

To obtain an expression for the capillary pressure P_σ we write the interface as

$$y=a+\xi(x)\ , \tag{3.18}$$

where $\xi(x)$ is the displacement of the interface at point x given by

$$\xi(x)=\int dt\, v_y(x,y=a-)=\frac{-ik}{\omega}\psi(x,y=a-)\ . \tag{3.19}$$

The capillary pressure P_σ, then, in $d=2$ is

$$P_\sigma=-\sigma\frac{\partial^2\xi}{\partial x^2}=\sigma k^2\xi \tag{3.20}$$

[see Eq. (2.3) above and Eq. (3.24) below]. With (3.18) and (3.20) the boundary condition (3.14) gives

$$B_1'\left[4\frac{\eta}{\eta'}k^2\sinh(ka)+2\omega\frac{\rho}{\eta'}\sinh(ka)+2\sigma\frac{(ka)^2}{a^2}\frac{k}{\omega\eta'}\cosh(ka)\right]+B_2'\left[4\frac{\eta}{\eta'}kK_1\sinh(K_1a)+2\sigma\frac{(ka)^2}{a^2}\frac{k}{\omega\eta'}\cosh(K_1a)\right]+B_1\left[\frac{\omega\rho'}{\eta'}+2k^2\right]e^{-ka}+B_22kK_1'e^{-ka}=0\ . \tag{3.21}$$

Equations (3.10), (3.11), (3.13), and (3.21) determine the dispersion relation $\omega(k)$. This is a nontrivial transcendental equation for ω. Following Tomotika we take the large viscosity limit. Effectively, this means we will not obtain the standard results of capillary wave dispersion or capillary wave damping. Straightforward algebra gives as a final result

$$\omega = -\frac{\sigma}{2\eta a}\Phi_2(\eta/\eta', ka) , \tag{3.22}$$

where

$$\Phi_2^{-1} = \frac{\eta/\eta'}{ka}\left[1+\frac{\eta}{\eta'}\tanh(ka)\right] - \frac{1}{\cosh^2(ka)}\left[1-\frac{ka}{\frac{\eta}{\eta'}\cosh^2(ka)+\sinh(ka)\cosh(ka)+ka}\right] . \tag{3.23}$$

The function Φ_2 is positive definite. Thus, there are *no* wave numbers for which the system becomes unstable under an infinitesimal perturbation. The main difference between Eq. (3.22) and the corresponding $d=3$ result [Eq. (2.5)] involves the factor $(1-k^2a^2)$. This can be traced back to the expression for the capillary pressure [Eqs. (2.3) and (3.20)]: for a cylinder,

$$P_\sigma = \frac{\sigma}{a^2}\xi(k^2a^2-1) . \tag{3.24}$$

The k^2a^2 term comes from the circular cross section of the tube which is absent in $d=2$. In three dimensions, it is the competition between the two terms enclosed in the parentheses in Eq. (3.24) which leads to the instability. In two dimensions, fluctuations in the linear boundaries increase the curvature (and thus the interfacial free energy). Therefore there is no instability.

IV. GROWTH MECHANISMS IN TWO-DIMENSIONAL PHASE SEPARATION

Having obtained the result for the strip geometry, we now speculate on possible mechanisms for phase separation in two-dimensional binary fluids. We first consider dilute mixtures. After formation of domains, they grow initially by droplet coalescence. From dimensional analysis, the analog of Stoke's formula[15] in $d=2$ for the "bare" diffusion constant gives[26,27] $D \sim k_B T/\eta$, where k_B is Boltzmann's constant and T is temperature, which leads to

$$R^2 \sim (k_B T/\eta)t , \tag{4.1}$$

i.e., $R \sim t^{1/2}$. Equation (4.1) should give the dominant time behavior, up to possible logarithmic corrections.[25,27] This will be followed by an evaporation-condensation mechanism. The analog of the $d=3$ Lifshitz-Slyozov result[8] for two dimensions in somewhat difficult to obtain. For example, one requires the solution for the concentration c given by $\nabla^2 c=0$. A consistent treatment of the logarithmic solution of this equation requires the introduction of a large length scale cutoff. A detailed analysis has been given recently by Marqusee.[28] In any case, the time dependence of the evaporation-condensation process remains $R \sim t^{1/3}$ in two dimensions. This picture for $d=2$ dilute mixtures is quantitatively different from $d=3$, where both diffusive coalescence and evaporation condensation follow a $t^{1/3}$ behavior.

For concentrated mixtures, we expect a qualitative difference from $d=3$ phase separation. The linear stability analysis of Sec. III suggests that the tubular necking-down mechanism is inoperative in two dimensions. Thus it appears that the interconnected structure will coarsen initially and break up by interfacial diffusion, which follows a $t^{1/2}$ mechanism as in Eq. (4.1). Subsequently, evolution proceeds by droplet coalescence, which would again follow a $t^{1/2}$ behavior as in Eq. (4.1). (We would expect $d=2$ geometries to be chosen so that gravity plays no role.) Of course, the presence of large-amplitude fluctuations in the interconnected structure, due to the amplification of thermal fluctuations following the quench, could have unforseen effects, and other growth mechanisms could become important. Also, experiments would probably involve suspended monolayers, which could have different properties than the $d=2$ binary fluid we have discussed. In any case, the scenario we have described above should be a useful guide to experimentalists.

These results should also be valid for a liquid-vapor transition under certain conditions. In the late stages of evolution the system has reached local thermal equilibrium and the density has relaxed to near-equilibrium values in well-defined domains. Our analysis of the velocity field of an incompressible fluid again seems appropriate. Koch, Desai, and Abraham[14] have conducted a molecular-dynamics simulation of phase separation in a liquid-vapor system, at its critical concentration. Their simulations find no crossover to $R \sim t$ behavior, which is consistent with the scenario we have outlined above. A discussion of other growth mechanisms applicable to a liquid-vapor system is given in their paper.[29]

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[*] Permanent address: Departamento de Física Teorica, Universidad de Barcelona, Diagonal 647, 08028 Barcelona 28, Spain.

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