Hydrodynamic effects in phase separation: An explicit solution approach

Mariapia Riva* and Vincenzo Gino Benza[†]

*Dipartimento di Fisica, Università degli Studi di Milano, INFM, Unità di Milano,
and INFN, Sezione di Milano, via Celoria 16, 20133 Milano, Italy*

We study the influence of hydrodynamic modes on phase separation by modeling pure-phase domains with kinks. The velocity field, determined adiabatically in terms of the thermodynamic source, describes a steady state compressible flow. We consider spherical, columnar, and disklike domains. In the former two cases, we obtain crossover from the $\dot{R} \sim 1/R^2$ law to the $R \sim t$ law as in the incompressible case. The growth of columnar domains is dominated by hydrodynamics, while for spherical domains we find competition between hydrodynamics and particle diffusion: this competition determines a threshold in size for the fluctuations, i.e., only large enough fluctuations can initiate the spinodal decomposition. We finally study disklike domains in a quasi-two-dimensional system and obtain, for small radii, the growth law $\dot{R} \sim 1/[R^3 \ln(\Lambda/R)]$, while for large radii $\dot{R} \sim 1/[R^2 \ln(\Lambda/R)]$. We find that stable microdomains can be formed, whose size is fixed by the thickness of the lamella and by the parameters of the fluid (e.g., viscosity, transport coefficient, etc.). [S1063-651X(97)02712-8]

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I. INTRODUCTION

The influence of hydrodynamics on phase separation in binary mixtures has been the object of numerical [1–5] as well as analytical works based on dimensional and scaling analysis [6,7]. We approach the problem in terms of explicit analytic solutions of the dynamics of the order parameter, representing the concentration, and the fluid velocity. Our approach is an extension of the Lifshitz-Slyozov analysis [8] to include the hydrodynamic contribution: we obtain an evolution equation for the size of an isolated pure-phase domain. The velocity field is determined in the adiabatic approximation, meaning that the viscous relaxation occurs over short times with respect to the times scales of phase separation. In this way we disregard memory effects; in general the velocity of the fluid is consistent with the stationary flow imposed by the quasistatic chemical potential. The dynamics of the interface results from the competition between the diffusion-dominated dynamics (single type pure-phase molecules, say B, diffuse within the melt towards the domain) and the global motion of the melt, described by the hydrodynamic modes. We consider an off-critical quench, i.e., the melt is assumed to be close to the equilibrium concentration of phase A: the free energy density is then approximated with a quadratic term. It will be shown that as a consequence the transverse part of the velocity decouples from the chemical potential. Our solutions refer to potential, compressible flows; we discuss in the following how the kinetic ordering is influenced by relaxing the incompressibility condition.

The paper is organized as follows. In Sec. II we introduce the basic equations and discuss the separation of the Navier-Stokes equations in the static limit in longitudinal and transversal parts. In Sec. III we consider a spherical domain and show that, in the absence of a forcing term at the boundary, a characteristic radius separates collapsing domains from expanding ones. The crossover from a Lifshitz-Slyozov behavior to the hydrodynamic behavior is also obtained. In Sec. IV we study a columnar defect; in the limit of infinite system only the hydrodynamic contribution survives and a stable time-dependent solution is found. This solution is sustained only by the forcing at the boundary: in the absence of forcing all columnar defects grow with a linear law. In Sec. V we consider a disk in a (quasi-two-dimensional) laminar system: we find here a deviation from the Lifshitz-Slyozov growth for small domains (a slower growth) and for large domains a logarithmic correction to the Lifshitz-Slyozov law. More interestingly perhaps, we determine a characteristic radius, dependent on the thickness of the lamella and on the viscosity, where the domains stabilize. If one disregards the interaction between the domains, the system separates into a pattern of microdomains. Section VI is dedicated to some conclusions and to comparison with previous works.

II. BASIC EQUATIONS

Phase-separation dynamics of binary fluids will be studied here in terms of explicit solutions, based on kinklike modeling of the pure-phase domains.

Our starting point is the so-called H model [9], in its deterministic version:

$$\rho_0 \left[\frac{\partial \vec{v}}{\partial t} + (\vec{v} \cdot \vec{\nabla}) \vec{v} \right] = -\vec{\nabla} p + \eta \Delta \vec{v} + \left(\zeta + \frac{1}{3} \eta \right) \vec{\nabla} (\vec{\nabla} \cdot \vec{v})$$
$$- \lambda \varphi \vec{\nabla} \mu, \tag{1}$$

$$\frac{\partial}{\partial t} \varphi + \lambda \vec{\nabla} \cdot (\varphi \vec{v}) = M \Delta \mu. $$

*Electronic address: Mariapia.Riva@mi.infn.it
[†]Electronic address: Benza@mi.infn.it

Here $\varphi(\vec{x},t)$ is a scalar field describing the concentration of one of the two components, $\vec{v}(\vec{x},t)$ is the fluid velocity, ρ_0 the average mass density, $p(\vec{x},t)$ the pressure, and $\mu[\varphi]$ the chemical potential. The coefficients η and ζ are the shear and bulk viscosity, λ the coupling of the concentration with the velocity, and M a transport coefficient, here assumed to be constant. We study the scaling behavior during phase separation in the deterministic case: it was numerically shown that noise is irrelevant in this regime in $d=2$ and $d=3$ [3]. To the best of our knowledge, the noise term generates a peculiar scaling in monolayers in the coalescence regime [7], which is far from the present approach.

We examine time scales of the order of the phase-separation dynamics and large with respect to the transient viscous relaxation times of the velocity field: in other words, we study the system in the adiabatic approximation $(d/dt)\vec{v}=\vec{0}$, where the velocity ''instantaneously'' follows the dynamics of the order parameter. This approximation is particularly reasonable in proximity to the phase separation, where the system undergoes critical slowing down, but here we will use it in studying the evolution after quenching below the transition temperature. The experimental relevance of this adiabatic regime has been discussed by Koga and Kawasaki [4], in their numerical study of spinodal decomposition.

It is useful to project the residual Navier-Stokes equation into the transverse and longitudinal components $(\vec{v}=\vec{v}_T+\vec{v}_L, \vec{\nabla}\cdot\vec{v}_T=0, \vec{\nabla}\times\vec{v}_L=\vec{0})$:

$$-\vec{\nabla}p+\eta^*\Delta\vec{v}_L-\lambda(\varphi\vec{\nabla}\mu)_L=\vec{0},$$

$$\eta\Delta\vec{v}_T-\lambda(\varphi\vec{\nabla}\mu)_T=\vec{0},$$

where $\eta^*=\frac{4}{3}\eta+\zeta$. We point out that whenever the chemical potential μ is a polynomial in φ, $\mu=\Sigma_{i=1}^k a_i\varphi^i$ (this is a reasonable approximation when the melt is close to an equilibrium configuration, so that one can disregard in μ the kinetic term), the two equations decouple:

$$\eta^*\Delta\vec{v}_L=\vec{\nabla}p+\lambda\vec{\nabla}\left(\sum_{i=1}^k\frac{i}{i+1}a_i\varphi^{i+1}\right),$$

$$\eta\Delta\vec{v}_T=\vec{0}.$$

Notice that if one enforces the incompressibility condition $\vec{\nabla}\cdot\vec{v}=0$, one simply ends up with a solution for the pressure as a function of the potential, while $\vec{v}_T$ is completely independent of the chemical potential. In such a case the problem reduces to one for a scalar in a given external static velocity field with appropriate boundary conditions at infinity and at the interface. Here, we disregard the variation in space of pressure and include the effect of the source in the solution for the irrotational field $\vec{v}_L$, which will be uniquely determined by imposing $\vec{v}_L=\vec{0}$ on the boundary of the system.

For more general μ one can again determine the velocity field by simply inverting the Laplacian; indeed, by setting $\vec{u}=\eta^*\vec{v}_L+\eta\vec{v}_T$ one obtains

$$\Delta\vec{u}=\lambda\varphi\vec{\nabla}\mu+\vec{\nabla}p.$$

Starting from the solutions of $\Delta\mu=0$, valid in the static limit, we will assume kinklike geometries and determine a dynamical equation for the size of such kinks in a quasistatic approach. The procedure follows the lines of the derivation given by Langer [10] of the seminal paper by Lifshitz and Slyozov [8].

III. SPHERICAL SYMMETRY

We first consider a spherical domain having radius R in a pure phase, say B, surrounded by the mixed phase, assumed to be close to the A-phase equilibrium concentration c_α. Recalling that $\mu(\vec{x},t)=\delta F[\varphi]/\delta\varphi(\vec{x},t)$, we assume $\mu(\vec{x},t)=V''(c_\alpha)\varphi(\vec{x},t)$, where $V(\varphi)$ is the potential contribution to the free energy density.

The solution of $\Delta\mu=0$ is given, for $r>R$, by

$$\varphi(r)=a_s\frac{R}{r}+b_s, \tag{2}$$

with boundary conditions

$$\varphi=\begin{cases} c_\alpha+\delta c_\alpha & \text{if } r=R \\ c_\alpha+\delta c_\infty & \text{if } r=\Lambda, \end{cases}$$

where Λ is the radius of the system. The quantity δc_α is the excess concentration at the boundary of the domain, given by the Gibbs-Thompson relation:

$$\delta c_\alpha=2\frac{d_0}{R}\Delta c, \tag{3}$$

where $\Delta c=c_\beta-c_\alpha$, $d_0=\sigma\widetilde{\chi}/(\Delta c)^2$, σ being the surface tension and $\widetilde{\chi}$ the susceptibility. From the boundary conditions one determines the coefficients of the static solution:

$$a_s=\frac{\Lambda}{\Lambda-R}(\delta c_\alpha-\delta c_\infty), \tag{4}$$

$$b_s=c_\alpha+\frac{\Lambda}{\Lambda-R}\delta c_\infty-\frac{R}{\Lambda-R}\delta c_\alpha. \tag{5}$$

We solve the equations for $\vec{v}$ under the assumptions $\vec{\nabla}p=\vec{0}$ and $\vec{v}_T=\vec{0}$:

$$\Delta\vec{v}=\lambda\vec{f},$$

$$\vec{f}=\frac{1}{\eta^*}\varphi\vec{\nabla}\mu.$$

Notice that $\varphi(r)$ parametrically depends on the radius of the sphere R. From the symmetry and the equation $\Delta\mu=0$, one has

$$\vec{f}(\vec{r})=f(r)\vec{a}_r, \quad \vec{v}(\vec{r})=v(r)\vec{a}_r, \tag{6}$$

$$f(r) = \Theta(r-R) \frac{R^2}{\eta *} [\mu(r)]'_{r=R^+} \frac{1}{r^2} \varphi(r),$$

where $\vec{a}_r$ is the unit radial vector at the point $\vec{r}$ and $\Theta(x)$ is the Heaviside function.

We solve for $v(r)$ with the boundary conditions

$$v(r=\Lambda)=0, \quad G_s(r,\theta,\phi;r'=\Lambda,\theta',\phi')=0, \quad (7)$$

G_s being the Green function of the Laplacian operator in spherical coordinates.

On the surface of the domain we obtain

$$v(R) = -\frac{\lambda}{3\eta *} [\mu(\rho)]'_{\rho=R^+} R^2 \left[b_s + \frac{a_s}{2} - \frac{3}{2} \frac{R}{\Lambda} \left(b_s + a_s \frac{R}{\Lambda} \right) \right. $$
$$\left. + \frac{R^3}{\Lambda^3} \left(\frac{b_s}{2} + a_s \right) \right]. \quad (8)$$

We then integrate the equation of motion for φ over a region S_ϵ, covering the interface between $R-\epsilon$ and $R+\epsilon$. The quasistatic approach amounts to assuming that φ is time dependent according to $\varphi_{R(t)}(r) = \varphi(r-R(t))$ $[r \neq R(t)]$.

Following Refs. [10,11], we approximate $\varphi_{R(t)}(r)$ with the equilibrium value c_β inside the bubble and with the static solution outside it $[\varphi_{R(t)}(r)=\varphi(r)$ for $r>R+\epsilon]$. The average of $\dot{\varphi}_R$ becomes

$$\langle \dot{\varphi}_R \rangle_{S_\epsilon} = -\dot{R} \langle \varphi_R' \rangle_{S_\epsilon} \simeq 4\pi R^2 \dot{R} \Delta c.$$

Similarly we get

$$\langle \vec{\nabla} \cdot (\varphi_R \vec{v}) \rangle_{S_\epsilon} \simeq -4\pi R^2 v(R) \Delta c.$$

We point out that the contribution from $\langle \varphi_R(\vec{\nabla} \cdot \vec{v}) \rangle_{S_\epsilon}$ can be disregarded in the previous average; in fact, since $\vec{\nabla} \cdot \vec{v}$ is continuous across the interface, this term is $O(\epsilon)$.

Upon averaging the right side term of the equation for the order parameter we obtain

$$\dot{R} = \frac{M}{\Delta c} [\mu(r)]'_{r=R^+} + \lambda v(R), \quad (9)$$

where

$$[\mu(r)]'_{r=R^+} = V''(c_\alpha) \frac{1}{R} (\delta c_\infty - \delta c_\alpha) \frac{\Lambda}{\Lambda-R}. \quad (10)$$

In the right-hand side of Eq. (9) the first term represents (apart from the sign) the velocity of β particles entering the bubble, driven by diffusion. The second term is the global velocity of the mixed fluid, evaluated at the interface; for infinite systems, it is opposite to the gradient of the chemical potential [see Eq. (8)], as it must be, since it describes the corresponding stationary flux. Schematically we have

$$\dot{R} = -(v_\beta)_{\text{chem}} + \lambda v_{\text{hydr}}. \quad (11)$$

Notice that $(v_\beta)_{\text{chem}}$ is always opposite to the gradient of μ: hence the bubble growth is regulated by the velocity of β particles relative to the background hydrodynamic flow.

The previous equations can be written as

$$\dot{R} = \frac{M_H}{\Delta c} [\mu(r)]'_{r=R^+},$$

$$M_H = M - \lambda^2 \frac{\Delta c}{3\eta *} R^2 \left[b_s + \frac{a_s}{2} - \frac{3}{2} \frac{R}{\Lambda} \left(b_s + a_s \frac{R}{\Lambda} \right) \right.$$
$$\left. + \frac{R^3}{\Lambda^3} \left(\frac{b_s}{2} + a_s \right) \right]. \quad (12)$$

It appears that the hydrodynamic effects are totally included in an additional contribution to the transport coefficient.

The limit $\lambda \gg 1$ corresponds to a regime dominated by hydrodynamics: we then expect to recover the scaling $R \sim t$ [6]. In particular, as numerically shown in Ref. [2], upon increasing λ one should have a crossover from a diffusion-dominated growth $R \sim t^{1/3}$ to the hydrodynamic linear growth $R \sim t$.

For an infinite sample ($\Lambda \to \infty$) one obtains

$$\dot{R} = 2d_0 V''(c_\alpha) \frac{1}{R} \left(\frac{1}{R_{\text{cr}}} - \frac{1}{R} \right) \left[M - \lambda^2 \frac{(\Delta c)^2}{3\eta *} d_0 R^2 \right.$$
$$\times \left. \left(\frac{c_\alpha}{\Delta c} \frac{1}{d_0} + \frac{1}{R} + \frac{1}{R_{\text{cr}}} \right) \right], \quad (13)$$

where $R_{\text{cr}} = 2\Delta c / \delta c_\infty d_0$. Clearly Eq. (13) reduces to the Lifshitz-Slyozov equation [8] for $\lambda=0$. It is then obvious that starting from $\lambda=0$ one goes from a $R \sim t^{1/3}$ behavior to the linear behavior for large enough λ.

For the previously mentioned reasons, here the effective transport coefficient M_H has no definite sign. In particular at $R=\bar{R}$ we have $M_H=0$:

$$\bar{R} = \frac{\sqrt{\frac{1}{4} \left(\frac{\Delta c}{c_\alpha} \right)^2 d_0^2 + \left(1 + \frac{\Delta c}{c_\alpha} \frac{d_0}{R_{\text{cr}}} \right) l_s^2} - \frac{1}{2} \frac{\Delta c}{c_\alpha} d_0}{1 + \frac{\Delta c}{c_\alpha} \frac{d_0}{R_{\text{cr}}}}, \quad (14)$$

where $l_s = \sqrt{3M\eta * / (\lambda^2 c_\alpha \Delta c)}$. Notice that $\bar{R}$ increases with R_{cr} and has a finite limit for $R_{\text{cr}} \to \infty$. The dynamics is characterized by the two stationary solutions $\bar{R}$ and R_{cr}, the smallest one being always unstable.

Whenever $R < \bar{R} < R_{\text{cr}}$ the pure-phase fluctuations of size R are suppressed, while for $\bar{R} < R < R_{\text{cr}}$, they tend to grow; in particular, when $\bar{R} \ll R < R_{\text{cr}}$ one has

$$R \sim \left(\frac{2}{3} \lambda^2 \frac{\sigma}{\eta *} \frac{c_\alpha}{\Delta c} \right) t. \quad (15)$$

This is consistent with the result obtained by Siggia [6], and with successive scaling analyses (Refs. [2,7]) as well as numerical estimates [1]. These scaling results in $d=3$ are well known, but we point out that here they are obtained from an

analytic solution based on a radial, compressible flow over a compact domain, at odds with the standard arguments, based on transversal surface waves on extended domains.

Let us summarize the case $\bar{R} \ll R_{cr}$: when $R > \bar{R}$, $R \simeq \bar{R}$, one has $R \sim t^{1/3}$, while when $\bar{R} \lesssim R < R_{cr}$, $R \simeq R_{cr}$, one has $R \sim t$. Similarly, in the regime $R_{cr} \ll \bar{R}$ one finds for $R_{cr} < R < \bar{R}$ a growth crossover from $R \sim t^{1/3}$ to $R \sim t$; this regime can be reached by a strong forcing at the boundary and for large enough $M \eta^* / \lambda^2$.

IV. CYLINDRICAL SYMMETRY

We examine now the dynamics of a cylindrical domain having radius R, under the same physical conditions of the previous case.

The solution of $\Delta \mu = 0$ is, for $\rho > R$,

$$\varphi(\rho) = \frac{a_c}{\ln(\Lambda/R)} \ln \frac{\Lambda}{\rho} + b_c, \qquad (16)$$

where Λ is now the horizontal radius of the system, which in the z direction extends to infinity. The excess concentration δc_α at the interface $\rho = R$ can be determined as for the Gibbs-Thompson relation:

$$\delta c_\alpha = \frac{d_0}{R} \Delta c. \qquad (17)$$

From the boundary conditions at $\rho = R$ and $\rho = \Lambda$ one obtains the coefficients a_c and b_c:

$$a_c = \delta c_\alpha - \delta c_\infty, \quad b_c = c_\alpha + \delta c_\infty. \qquad (18)$$

We determine $\vec{v}$ from the equation $\Delta \vec{v} = \lambda \vec{f}$ by letting

$$\vec{f}(\vec{r}) = f(\rho) \vec{a}_\rho, \quad \vec{v}(\vec{r}) = v(\rho) \vec{a}_\rho,$$

$$f(\rho) = \Theta(\rho - R) \frac{R}{\eta^*} [\mu(\rho)]'_{\rho = R^+} \frac{1}{\rho} \varphi(\rho),$$

$$v(\rho = \Lambda) = 0, \qquad (19)$$

where $\vec{a}_\rho$ is the unit radial vector. The result at the interface is

$$v(R) = -\frac{\lambda R^2}{2 \eta^*} [\mu(\rho)]'_{R^+} \left[\ln \frac{\Lambda}{R} \left(b_c + \frac{a_c}{2} \right) - \frac{b_c}{2} \right.$$

$$\left. + \frac{R^2}{2 \Lambda^2} (b_c + a_c) - \frac{a_c}{4 \ln(\Lambda/R)} \left(1 - \frac{R^2}{\Lambda^2} \right) \right]. \qquad (20)$$

As in the previous case we average over a region C_ϵ covering the interface. We have

$$\langle \varphi_R \rangle_{C_\epsilon} = -\dot{R} \langle \varphi_R' \rangle_{C_\epsilon} \simeq 2 \pi \dot{R} R \Delta c \int dz$$

and similarly

$$\langle \vec{\nabla} \cdot (\varphi_R \vec{v}) \rangle_{C_\epsilon} \simeq -2 \pi R v(R) \Delta c \int dz.$$

Notice that the z dependence factors out both from the Lagrangian derivative and from the chemical source term.

It turns out that again $\vec{\nabla} \cdot \vec{v}$ is continuous across the interface, so that the incompressibility condition does not modify the equation for $R(t)$, which has the form (12), with

$$[\mu(\rho)]'_{\rho = R^+} = V''(c_\alpha) \frac{1}{R} (\delta c_\infty - \delta c_\alpha) \frac{1}{\ln(\Lambda/R)} \qquad (21)$$

and

$$M_H = M - \lambda^2 \frac{\Delta c}{2 \eta^*} R^2 \left[\ln \frac{\Lambda}{R} \left(b_c + \frac{a_c}{2} \right) - \frac{b_c}{2} + \frac{R^2}{2 \Lambda^2} (b_c + a_c) \right.$$

$$\left. - \frac{a_c}{4} \frac{1}{\ln(\Lambda/R)} \left(1 - \frac{R^2}{\Lambda^2} \right) \right]. \qquad (22)$$

It is easily verified that in the purely two-dimensional case, in the absence of hydrodynamic effects, one obtains exactly this equation with $\lambda = 0$, provided that Λ is finite; the limit $\Lambda \to \infty$ gives $\dot{R} = 0$. Notice that instead in the present context the limit $\Lambda \to \infty$ gives a nontrivial result:

$$\dot{R} = -\lambda^2 \frac{V''(c_\alpha)}{4 \eta^*} (\Delta c)^2 d_0^2 \left(\frac{1}{R_{cr}} - \frac{1}{R} \right) R \left(2 \frac{c_\alpha}{\Delta c} \frac{1}{d_0} + \frac{1}{R} + \frac{1}{R_{cr}} \right), \qquad (23)$$

with $R_{cr} = (\Delta c / \delta c_\infty) d_0$. Hence for the infinite system the hydrodynamic modes are the only surviving source of the dynamics.

We point out that the stability is modified with respect to the Lifshitz-Slyozov case, as well as the scaling behavior. A comprehensive description of the time evolution of R_{cr} would involve a statistical analysis for an assembly of columnar domains, including a global conservation law. We limit the treatment to the absence of forcing at the boundary, corresponding to $R_{cr} \to \infty$. In this case, for finite R, one obtains

$$R \sim \left(\frac{1}{2} \lambda^2 \frac{\sigma}{\eta^*} \frac{c_\alpha}{\Delta c} \right) t. \qquad (24)$$

This confirms known results, but, as previously pointed out, the mechanism is based on longitudinal excitations, while in Ref. [6] a pressure gradient parallel to the axis of the tube was assumed, involving transverse modes.

In a finite system, when R is small enough, there is a regime in which the hydrodynamic effects are dominated by particle diffusion; we obtain, in such a situation,

$$\dot{R} \sim \frac{1}{R^2 \ln(\Lambda/R)}. \qquad (25)$$

Consistently with the Lifshitz-Slyozov [8] law $R \sim t^{1/3}$, at larger R we get instead a crossover to a linear growth.

V. QUASI-TWO-DIMENSIONAL CASE

We consider the dynamics of a pure-phase disk in a cylindrical system with radius Λ and vertical thickness $l \ll \Lambda$.

While in the previous case the limit $\Lambda \to \infty$ gave a nonzero hydrodynamic velocity, here both the chemical velocity and the hydrodynamic velocity tend to zero whenever $\Lambda \to \infty$ with finite l.

We start with the static solution $\varphi(\rho)$ given by Eq. (15), and we take the Green function to be zero together with the hydrodynamic velocity on the boundary. The Green function $G_c(\vec{x}, \vec{x}'; l)$ is given by [12]

$$
G_c(\vec{x}', \vec{x}; l) = -\frac{4}{\pi^3 l \Lambda^2} \sum_{m=0}^{\infty} \sum_{n,s=1}^{\infty} \frac{1}{\epsilon_m} \frac{\cos m(\theta - \theta')}{J_{m+1}^2(\gamma_{m,s}) \left[\left(\frac{n}{l}\right)^2 + \left(\frac{\gamma_{m,s}}{\pi \Lambda}\right)^2 \right]} \sin\left(\frac{\pi n z}{l}\right) \sin\left(\frac{\pi n z'}{l}\right) J_m\left(\frac{\gamma_{m,s} \rho}{\Lambda}\right) J_m\left(\frac{\gamma_{m,s} \rho'}{\Lambda}\right),
$$

(26)

where $\epsilon_0 = 2$, $\epsilon_m = 1$ if $m \neq 0$, and $\gamma_{m,s}$ are the zeroes of the Bessel function J_m.

Upon letting $\vec{v}(\vec{x}) = v(\rho, z) \vec{a}_\rho$ one has

$$
v(\rho, z) = -\frac{4\lambda}{\pi^3 l \Lambda^2} \int_0^l dz' \int_0^\Lambda d\rho' \rho' \sum_{n,s=1}^{\infty} \frac{1}{J_2^2(\gamma_{1,s}) \left[\left(\frac{n}{l}\right)^2 + \left(\frac{\gamma_{1,s}}{\pi \Lambda}\right)^2 \right]} \sin\left(\frac{\pi n z}{l}\right) \sin\left(\frac{\pi n z'}{l}\right) J_1\left(\frac{\gamma_{1,s} \rho}{\Lambda}\right) J_1\left(\frac{\gamma_{1,s} \rho'}{\Lambda}\right) f(\rho').
$$

(27)

The average $V(R) = (1/l) \int_0^l dz v(R, z)$ can be written in the form

$$
V(R) = -\frac{16}{\pi^4 \Lambda} \frac{\lambda}{\eta^*} R[\mu(\rho)]_{R^+}' \int_{R/\Lambda}^1 dy \sum_n{}' \frac{1}{n^2} S\left(n, y; \frac{R}{\Lambda}\right)
$$

$$
\times \left[a_c \frac{\ln y}{\ln(R/\Lambda)} + b_c \right],
$$

(28)

where the prime means that the sum extends over odd values of n, and S has the following form:

$$
S\left(n, y; \frac{R}{\Lambda}\right) = \sum_{p=1}^{\infty} \frac{J_1(\gamma_{1,p} R/\Lambda) J_1(\gamma_{1,p} y)}{J_2^2(\gamma_{1,p}) \left[\left(\frac{n}{l}\right)^2 + \left(\frac{\gamma_{1,p}}{\pi \Lambda}\right)^2 \right]}.
$$

(29)

It is found that $S(n, y; R/\Lambda)$ can be exactly summed [13],

$$
S\left(n, y; \frac{R}{\Lambda}\right) = -\Lambda^2 \frac{\pi^3}{4} \frac{J_1(i \pi (\Lambda/l) n (R/\Lambda))}{J_1(i \pi (\Lambda/l) n)}
$$

$$
\times \left[J_1\left(i \pi \frac{\Lambda}{l} n\right) Y_1\left(i \pi \frac{\Lambda}{l} n y\right) \right.
$$

$$
\left. - J_1\left(i \pi \frac{\Lambda}{l} n y\right) Y_1\left(i \pi \frac{\Lambda}{l} n\right) \right].
$$

(30)

In the limit $l/\Lambda \ll 1$ one approximates the Bessel functions with their asymptotic behavior; in Eq. (28) we first integrate in dy and then sum over n, thus finally obtaining

$$
V(R) = -\frac{\lambda}{24\eta^*} l^2 [\mu(\rho)]_{R^+}' (a_c + b_c).
$$

(31)

Hence the final result for the effective transport coefficient M_H is given by

$$
M_H = M - \lambda^2 \frac{(\Delta c)^2}{24\eta^*} l^2 \left(\frac{c_\alpha}{\Delta c} + \frac{d_0}{R} \right),
$$

(32)

meaning that a length $l_c = \sqrt{24 M \eta^* / (\lambda^2 c_\alpha \Delta c)}$ separates two regimes: (a) $l \geq l_c$, where $M_H < 0$ and $R_{\rm cr}$ is the unique (stable) zero of $\dot{R}$; (b) $l < l_c$, where $\dot{R}$ has two zeroes, $R_{\rm cr}$ and $\bar{R}$:

$$
\bar{R} = d_0 \frac{\Delta c}{c_\alpha} \frac{l^2}{l_c^2 - l^2}.
$$

(33)

An interesting situation holds in case (b), where the evolution equation has the form

$$
\dot{R} = M V''(c_\alpha) d_0^2 \frac{(\Delta c)^2}{c_\alpha} \frac{l^2}{l_c^2} \frac{1}{R} \left(\frac{1}{\bar{R}} - \frac{1}{R} \right) \left(\frac{1}{R_{\rm cr}} - \frac{1}{R} \right) \frac{1}{\ln(\Lambda/R)}.
$$

(34)

Independently of the relative magnitude of $R_{\rm cr}$ and $\bar{R}$, the largest zero is always unstable. As a consequence, the domains are growing in the two regions external to the roots.

From Eq. (34) it is found that whenever $R < R_{\rm cr}, \bar{R}$, the growth law is

$$
\dot{R} \sim \frac{1}{R^3 \ln(\Lambda/R)}.
$$

(35)

It is easily verified that, as $l \to 0$, the evolution equation (34) reduces to the two-dimensional case without hydrodynamic effects.

It should be noticed that, when $l < l_c$, Λ is finite and the system is not forced from the boundary ($\delta c_\infty = 0$), a stable configuration of pure phase disks with radius $\bar{R}$ is expected from Eq. (34); $\bar{R}$ depends on the scales of the system only through the thickness l and tends to zero as $\lambda \to 0$. This

microphase separation is sustained by hydrodynamics: pure-phase fluctuations would be damped in a static background under such conditions.

The flow given by Eq. (31) follows Darcy's law with the chemical potential in the role of an effective pressure, and a coefficient of permeability proportional to l^2 and to the concentration φ. In general the flow depends on the size of the system, while the chemical velocity is merely proportional to the pressure gradient: in particular, this implies that the dynamics of infinitely long tubular domains is only dependent on hydrodynamics [see Eq. (23)]. In the monolayer case, a stable disklike solution is made possible by the $1/R$ behavior of the concentration φ at the interface: this factor, multiplying the gradient of the chemical potential, makes $v_{\text{hydr}} > v_{\text{chem}}$ for small enough R ($R < \bar{R}$). In this regime the domain globally increases. When $R > \bar{R}$ the opposite occurs, so that the domain tends to shrink. This mechanism is inhibited in the spherical case since the $1/R$ behavior of φ is there multiplied by a $\sim R^2$ factor coming from the inverse Laplacian [see Eq. (12)]: as a result, the value of R corresponding to $v_{\text{hydr}} = v_{\text{chem}}$ is always unstable. Similarly for a thick film ($l > l_c$) the hydrodynamic flow is always larger than the chemical potential, so that qualitatively everything goes as in Sec. IV.

Whenever $\bar{R} < R_{\text{cr}}(0) < \infty$ the dynamics becomes very involved, and only within a statistical treatment can one determine the steady state: since $\bar{R}$ acts as an attractor, $R_{\text{cr}}(t)$ is not *a priori* allowed to grow indefinitely as in the Lifshitz-Slyozov case, which corresponds here to $\bar{R} = 0$. One can estimate the growth law for $\bar{R} < R_{\text{cr}} < R$ as

$$\dot{R} \sim \frac{1}{R^2 \ln(\Lambda/R)}.$$

This confirms the scaling law already obtained in the cylindrical case under similar conditions.

In the purely two-dimensional case, by analyzing the stability of a strip, San Miguel *et al.* [14] concluded that a linear growth $R \sim t$ cannot be found. They further pointed out that their result should not be taken for granted in the case of monolayers; as a matter of fact, in our analysis we confirm the absence of linear behavior. Unfortunately, since we assume $v = 0$ on the boundary, we cannot follow the hydrodynamical effects in the limit $l \to 0$.

VI. CONCLUSIONS

We determined time evolution equations for pure-phase domains, including the hydrodynamic effects. Our explicit solutions in the case of spherical or columnar domains confirm a crossover from the $R \sim t^{1/3}$ to the $R \sim t$ law. We also explicitly determine the coefficients of the linear law, which

turn out to differ only through a numerical factor in the two cases [see Eqs. (15) and (24)]. In the limit of infinite system, while with spherical symmetry both the diffusion-driven separation and the fluid velocity do contribute to the interface dynamics, in the cylindrical symmetry case only the latter one survives. A second relevant difference, related to the previous point, is that while in the cylindrical case $R = 0$ is always unstable, in the spherical case there is a characteristic threshold below which pure-phase fluctuations are depressed and above which they grow towards spinodal decomposition.

The case of a laminar system shows a peculiar behavior: first of all a linear growth is never allowed, but this does not mean that hydrodynamics is not relevant. The most striking effect is the possibility of microdomain separation, the size of such domains being determined by the thickness of the lamella, the viscosity, and the transport coefficient. A new scaling behavior is found for small domains, which grow according to the law $\dot{R} \sim 1/[R^3 \ln(\Lambda/R)]$. In his renormalization group (RG) theory of phase-ordering kinetics, Bray [7] found in the Hele-Shaw cell an $R \sim t^{1/4}$ behavior corresponding to the coalescence regime; we believe that the correspondence with our result [see Eq. (35)] is merely coincidental, since we are considering strictly deterministic equations and coalescence is not accessible within the present treatment. On the other hand, we find that large domains follow the Lifshitz-Slyozov behavior, apart from a logarithmic correction: $\dot{R} \sim 1/[R^2 \ln(\Lambda/R)]$. We point out that this behavior is consistent with Bray's RG theory, in that actually an $R \sim t^{1/3}$ growth is predicted for off-critical quench, when the domains stabilize into cylindrical symmetry. We stress that the behavior $R \sim t^{1/2}$ in $d = 2$, as found by San Miguel *et al.* [14] and confirmed by Bray [7], is out of reach within our context.

Our results were derived with irrotational, compressible velocity fields. In general relaxing the incompressibility condition does not seem to modify the scaling behavior. The main feature of our approach is to consider a steady state flow generated by a specific chemical potential source: this makes our treatment intrinsically nonlinear, so that the flow is not merely determined by the boundary conditions, but is forced by a given configuration φ. The physical context here includes phase separation in binary gases having a miscibility gap, such as helium-hydrogen, helium-nitrogen, and neon-xenon [5,15]. A steady state compressible flow can also be found in liquid motion through porous media, where the medium can both absorb and relax material.

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