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Effect of inertia on droplet growth in a fluid

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The effect of inertia on droplet growth in a d-dimensional (simple) fluid mixture is investigated. Four typical growth laws of average droplet radius are obtained: two conventional ones ($t^{1/d}$ and t) and two new ones ($t^{2/(d+2)}$ and $t^{2/3}$). The regions of the applicabilities of these growth laws are investigated. Far away from the critical point, or for earlier or later stages of phase separation, new growth laws ($t^{2/(d+2)}$ and $t^{2/3}$) are dominant. These new laws represent droplet growth in the case of high Reynolds numbers where the inertia of the fluid is important and the system may be turbulent.

I. INTRODUCTION

It is believed that a many-body system exhibits a certain universality when a relevant length scale, say L, of the system becomes of macroscopic order. For the phase separation of systems with conserved order parameters or of certain systems with nonconserved order parameters, the scaling law exists. An average quantity G as a function of wave number k and time t has a scaling form (homogeneous form)

$$G_k(t)=[L(t)]^y \widetilde{G}(kL(t)) , \tag{1.1}$$

where y is a constant. The most used quantity for G is the structure function $S_k(t)=\langle n_k(t)n_{-k}(t)\rangle$, where n_k is the Fourier component of the local number-density difference (of one species) (see Fig. 1). From (1.1) and a dimensional consideration the structure function is assumed to be scaled as

$$S_k(t)=n^2L^d\widetilde{S}(Lk) , \tag{1.2}$$

where n is the density difference of one species between coexisting phases (in the small region with volume L^d) and d is the dimensionality. Since the structure function (structure factor) S is related to the susceptibility or the compressibility, χ_k, by the relation $\chi_k=k_BTS_k$, the scaling assumption (1.1) leads to a well-known scaling relation in the static case. This gives a relation among three scaling exponents. Two of them remain unknown. In the case of phase separation n is assumed to be fixed constant independent of the length scale L. Therefore, one may determine only one exponent, say a:

$$L(t)\propto t^a . \tag{1.3}$$

In the case of the static scaling, the time t in (1.3) is replaced by the temperature measured from the critical temperature T_c or more suitably by its inverse. Lifshitz and Slyozov[1] were the first to show such a power-law behavior of the length scale. Binder and Stauffer[2] were the first to put forward the above scaling idea and to show the variety of the exponent a. The present author[3] then gave a formulation based on the scaling assumption (1.1) and also arrived at the power law as (1.3). Siggia[4] discussed the cluster growth in a phase-separating viscous fluid mixture from a phenomenological but a quantitative viewpoint. Kawasaki and Ohta considered the basic equation suitable to the dynamics of phase separation to be that for the interface of droplets.[5] Since Marro *et al.*[6] ascertained the scaling assumption of structure function through numerical simulation, many studies were devoted to this from theoretical and experimental viewpoints.[7] The exponent a may be given in a more general form as[8]

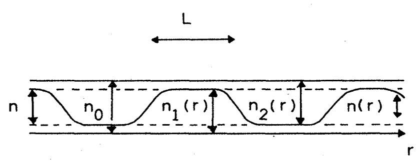
$$a=1/(d+\theta+\zeta-h) , \tag{1.4}$$

where θ is zero if the order parameter is not conserved, while it is 2 if the order parameter is conserved, and ζ and h are the scaling exponents associated with the mobility $M_k(t)$ and the energy $H_k(t)$:

$$M_k(t)=[L(t)]^{-\zeta}\widetilde{M}(Lk) , \tag{1.5}$$

$$H_k(t)=\langle(\delta F/\delta n_{-k})n_{-k}\rangle=[L(t)]^h\widetilde{H}(Lk) , \tag{1.6}$$

and where F is the free-energy functional. For the kinetic Ising system $\zeta=1-\theta/2$ and for fluid mixtures $\zeta=-2$. If the surface tension plays a role then $h=d-1$. On the other hand, if it does not play a role then $h=0$ at high



$$n(r)=n_1(r)-n_2(r) , \quad n_0=n_1(r)+n_2(r) ,$$

$$n_k=[n_1-n_2]_k \approx [n_1]_k \approx [n_2]_k ,$$

$$L^{d/2}n \approx [n_1-n_2]_{k=1/L}$$

FIG. 1. Quantities used in the text. Two species are assumed to be symmetric. $n_i(r)$ $(i=1,2)$ are densities of ith species, $n_0=n_1(r)+n_2(r)$ is the total number density, and $n(r)=n_1(r)-n_2(r)$, which may be an order parameter. A simplified expression n denotes a density difference of one species, say $i=1$, on the coexistence curve. The appearance of n in (1.2) is due to the estimate $(n_1-n_2)_{k=1/L}\approx L^{d/2}n$.

temperatures and $h=-\infty$ at low temperatures near $T=0$ (see, for instance, the first reference of Ref. 8).

Recent numerical simulations[9] for model systems with highly degenerated ground states exhibit "nonuniversal behaviors" of the droplet growth or the existence of the multiple "fixed-point values" of a. Very recently the same nonuniversal behavior was reported also for binary fluid mixture.[10] Such a nonuniversal behavior in the highly degenerate system may be explained as occurring due to the fact that the surface tension becomes ineffective[11] or becomes "intermittently" ineffective[12] for such systems for a geometrical reason.

So far all these growth laws are derived using the assumption that the droplet growth is described by a relaxational equation of motion for order parameters. In the fluid system this restricts the applicability of the above growth law to a narrow region near the critical point. The purpose of this paper is therefore to present a growth law for the binary fluid mixture in the whole region inside the coexistence curve. The key point of our theory is to add the contribution from the inertia term of the Navier-Stokes equation to the equation previously used. We then classify the situations into two cases where the inertia term is effective and where the dissipation term is effective. Each case is further classified into two cases where the surface tension is effective and where it is not effective. For the late stage of the droplet growth, the inertia term becomes effective. In such a case the exponent a can be given by a relation different from (1.4). Also, the fluid may be turbulent in such a case.

In Sec. II the basic equation is given. In Sec. III this equation is examined on the basis of the scaling assumption. In Sec. IV four growth laws for length scale L are obtained. Section V is devoted to the discussion and remarks.

II. BASIC EQUATION

Let us consider a two-phase coexistence. The phase separation proceeds by accompanying large scale flows of each phase. The average density n_0 of the fluid may be regarded as a constant independent of time. Therefore, it is sufficient for us to consider only the motion of one of coexisting phases. Let us start with the Navier-Stokes equation for the velocity U of one of coexisting phases:

$$\frac{D\vec{u}(\vec{r},t)}{Dt}=-\frac{1}{g_0}\vec{\nabla}p(\vec{r},t)+\nu\nabla^2\vec{u}(\vec{r},t)+\vec{f}^{(u)}(\vec{r},t)\,, \tag{2.1}$$

where D/Dt $(=d/dt+\vec{u}\cdot\vec{\nabla})$ denotes the time derivative along the fluid motion, g_0 the mass density of the fluid, p the local pressure in the fluid, and ν the kinetic viscosity. g_0 and ν may be assumed to be temperature and concentration independent. $\vec{f}^{(u)}$ is the fluctuating force, which is related to the kinetic viscosity through the fluctuation-dissipation theorem (see Appendix A). Let us assume that the phase separation proceeds isothermally. Then we can set $dp=n\,d\mu$, where μ is the chemical potential:

$$\mu=\frac{\delta F}{\delta n}\,. \tag{2.2}$$

Equation (2.1) can be rewritten as

$$\frac{D\vec{u}(\vec{r},t)}{Dt}=-\frac{1}{m}\vec{\nabla}\mu(\vec{r},t)+\nu\nabla^2\vec{u}(\vec{r},t)+\vec{f}^{(u)}(\vec{r},t)\,, \tag{2.3}$$

where $m=g_0/n=m_0n_0/n$ with m_0 and n_0 being a mass unit and an average density of the fluid, respectively. m_0 and n_0 are insensitive to the temperature while m is sensitive to the temperature. We now assume that the fluid collectively flows in the form of droplets or clusters. Therefore, the second term on the right-hand side of (2.3) may be replaced by

$$\nabla^2\rightarrow 1/(c_1L^2)\,, \tag{2.4}$$

where c_1 is a constant of the order unity. We obtain here the equation of motion for the order parameter $n(\vec{r},t)$. The velocity of the order parameter is also equal to $\vec{u}$ [notice that $n(\vec{r},t)=2n_1(\vec{r},t)-n_0$]. Substituting (2.4) into (2.3) and solving for $\vec{u}$, we have

$$\begin{aligned}\vec{j}(\vec{r},t)=n\vec{u}(\vec{r},t)=M&\left[-\vec{\nabla}\mu(\vec{r},t)-m\frac{D\vec{u}(\vec{r},t)}{dt}\right]\\&+m\vec{f}^{(u)}(\vec{r},t)\,,\end{aligned} \tag{2.5}$$

where

$$M=c_1nL^2/(m\nu) \tag{2.6}$$

is the mobility. $\vec{j}$ is the current density of the order parameter. Substituting (2.5) into the continuity equation for the order parameter,

$$\frac{dn(\vec{r},t)}{dt}+\vec{\nabla}\cdot\vec{j}(\vec{r},t)=0\,, \tag{2.7}$$

we obtain an equation for the order parameter:

$$\frac{dn(\vec{r},t)}{dt}=M\left[\nabla^2\mu(\vec{r},t)+m\vec{\nabla}\cdot\frac{D\vec{u}(\vec{r},t)}{Dt}\right]+f(\vec{r},t)\,, \tag{2.8}$$

where

$$f(\vec{r},t)=-Mm\vec{\nabla}\cdot\vec{f}^{(u)}(\vec{r},t) \tag{2.9}$$

is the fluctuating force of the order-parameter equation which is related to the mobility M through the fluctuation-dissipation relation (Appendix A)

$$\langle f_{\vec{k}}(t)f_{-\vec{k}}(t')\rangle=2k_BTMk^2\delta(t-t')\,. \tag{2.10}$$

Neglecting the second term, the inertia term, in large parentheses of (2.8) we obtain the equation previously used.[8] Another equation[13] can be obtained by rewriting the continuity equation (2.7) in the form

$$\frac{dn(\vec{r},t)}{dt}=\frac{Dn(\vec{r},t)}{Dt}-u(\vec{r},t)\cdot\vec{\nabla}n(\vec{r},t) \tag{2.7$'$}$$

and replacing the first term on the right-hand side of (2.7′) by a "nonhydrodynamic" term. We need several complicated procedures for the reduction of the second equation. Therefore, we do not use the second equation. Equation (2.8), along with (2.10), are our basic equations in the following discussion. Here we note that the left-hand side of (2.8) corresponds to the second term on the right-hand side of (2.3) and represents the energy dissipation. The second term on the right-hand side of (2.8) corresponds to the left-hand side of (2.3) and represents the inertia of the fluid motion. The remaining terms of both equations represent the driving force due to the inhomogeneity of the system.

It is convenient for us to transform (2.8) into the equation of motion for the structure function. A similar method as given in Appendixes A and B of Ref. 8 provides us with

$$\frac{dS_k(t)}{dt}+2I_k(t)=2Mk^2[k_BT-H_k(t)]\ , \tag{2.11}$$

where

$$I_k(t)=mM\,\mathrm{Re}\left\langle\left[i\vec{k}\cdot\frac{D\vec{u}_{\vec{k}}(t)}{Dt}\right]n_{-\vec{k}}(t)\right\rangle . \tag{2.12}$$

$H_k(t)$ in Ref. 8 is divided by k_BT, however. The first term and the second term on the left-hand side of (2.11) correspond to the dissipation term and the inertia term of the Navier-Stokes equation, respectively. The right-hand side of (2.11) represents the force terms, and corresponds to the fluctuating force and the pressure term of the Navier-Stokes equation. This equation will be used to obtain the growth law of the length scale L.

III. SCALING

If we set $I_k=0$, then (2.11) reduces to the equation previously used.[8] This equation leads to the relation (1.4) for a conserved order parameter n. We here discuss the scaling property of the term $I_k(t)$. Let us consider the Navier-Stokes equation (2.1). The left-hand side of this equation has a dimensionality Lt^{-2}. This term cannot be set as d^2L/dt^2, which may give a negative acceleration and therefore is unphysical. The reason for the inapplicability of the latter expression is as follows. The acceleration of a droplet continues for a time interval of the order of t for which the droplet moves a distance of the order of L. Then the droplet coalesces into larger ones. After that, the coalescing droplet is accelerated until it further coalesces into a larger one, and so on. Such accelerations should always be positive. Thus, those should be of the form Lt^{-2} owing to the scaling assumption. The acceleration d^2L/dt^2 represents the acceleration of a rigid body, and it is therefore negative if the velocity L/t is a decreasing function of t. Let us show here how we can connect a positive acceleration to a decreasing "velocity" of droplets. The kinetic energy per a unit mass at time t is of the order $(dL/dt)^2\approx(L/t)^2$. Let us estimate the total kinetic energy K_T which has been supplied to the fluid. Let ΔL be the increment of the average droplet radius in a time interval Δt. Then, due to the scaling assumption one may find that

$$\Delta L/\Delta t\approx dL/dt=aL/t\ .$$

Thus, it takes a time interval of the order t $(=L^{1/a})$ for a droplet radius to be increased by the amount of order L. This means that the time rate of the birth of a droplet with a radius $L(=t^a)$ is proportional to $1/t$. Thus, we find that

$$K_T\approx\int_0^t\left[\frac{dL}{dt'}\right]^2 t'^{-1}dt'\ . \tag{3.1}$$

Therefore, the acceleration of the fluid at time t is given by

$$\left[\frac{dL}{dt}\right]^{-1}\frac{dK_T}{dt}\approx\left[\frac{dL}{dt}\right]t^{-1}\approx Lt^{-2}\ \ (>0)\ . \tag{3.2}$$

The inertia term may be divided into two parts. One is the part which is directly coupled with the pressure term. The acceleration (3.2) is for such a part of the inertia term. The other is a part which is indirectly coupled with the pressure term through the nonlinear term of the Navier-Stokes equation. The energy integral (3.1) contains all these parts of the inertia term. In the present paper attention is paid only to the large scale motion (a growing or a frontier motion). The stationary or the residual motion may be important to the discussion of the energy cascade in a case of turbulence. This is not, however, the case which we are interested in in the present paper. Let us put

$$S_k(t)\,|_{k=L^{-1}}=c_2n^2L^d\ , \tag{3.3}$$

$$\frac{Du}{dt}\rightarrow a_2Lt^{-2}\ , \tag{3.4}$$

where c_2 is a constant coming from an integration for the Fourier transformation. a_2 and c_2 are of the order unity. From (3.3) and (3.4), (2.11) can be evaluated as

$$c_2n^2L^dt^{-1}+2a_2c_2mMnL^dt^{-2}=2ML^{-2}(k_BT-H_L)\ , \tag{3.5}$$

or by multiplying both sides by $L^{-d+1}/(2Mg_0)$ we obtain

$$c_2\nu/(2c_1Lt)+a_2c_2Lt^{-2}=(k_BT-H_L)/(g_0L^{d+1})\ . \tag{3.6}$$

The transformation from (3.5) into (3.6) is done so that (3.6) may have the same dimensionality as that of the Navier-Stokes equation.

IV. GROWTH LAWS

There are two main contributions to the energy H_L. The one is from a configurational entropy of droplets, $H^{(e)}$. The other is from the surface tension, $H^{(s)}$. These are scaled respectively as (see Ref. 8)

$$H^{(e)}=(1-a_0)k_BT,\ \ H^{(s)}=-a_1\sigma L^{d-1}\ , \tag{4.1}$$

where σ is the surface tension. Here the superscripts (e) and (s) respectively mean the entropy and the surface tension. Then the right-hand side of (3.6) is written as

$$P^{(e)}=a_0k_BT/(g_0L^{d+1}),\ \ P^{(s)}=a_1\sigma/(g_0L^2)\ , \tag{4.2}$$

corresponding to $H^{(e)}$ and $H^{(s)}$, respectively. Let us denote the first term (i.e., the dissipation term) and the second term (i.e., the inertia term) on the left-hand side of (3.6) respectively by D and I:

$$D=c_2\nu/(2c_1Lt),\ \ I=a_2c_2Lt^{-2}\ . \tag{4.3}$$

Let us define two quantities:

$$Q = P^{(e)}/P^{(s)}, \quad R = I/D \ . \tag{4.4}$$

Here R is the Reynolds number. Equation (3.6) is now rewritten as

$$D + I = P^{(e)} + P^{(s)} \ . \tag{3.6$'$}$$

Then four regions are considered, i.e., region I, $Q \ll 1$ and $R \ll 1$; region II, $Q \gg$ and $R \ll 1$; region III, $Q \ll 1$ and $R \gg 1$; and region IV, $Q \gg 1$ and $R \gg 1$. In each of four regions the value of the exponent a is given. For example, in the region I the exponent a is given by the equation $D = P^{(s)}$. This leads to $a = 1$ because of the first part of (4.3) and the second part of (4.2). Values of a in other regions are given in the same way. Let us introduce dimensionless quantities $\tilde{L}$ and $\tilde{t}$ defined by

$$\tilde{L} \equiv L/\xi, \quad \tilde{t} \equiv (\xi^2/D_T)^{-1} t \ , \tag{4.5}$$

where ξ is the thermal correlation length and D_T the thermal diffusion constant on the coexistence curve at the same temperature. The diffusion constant D_T is given in the present notation by (Appendix B)

$$D_T = c_1 k_B T / (c_2 \nu g_0 \xi^{d-2}) \ . \tag{4.6}$$

Then the two quantities defined by (4.4) are respectively written as

$$Q = C_1 \tilde{L}^{1-d}, \quad R = C_2 Z \tilde{L}^{2} \tilde{t}^{-1} \ , \tag{4.7}$$

where $C_1 = a_0 / a_1 e_1$, $C_2 = 2a_2 c_1$, and

$$Z = D_T / \nu \ , \tag{4.8}$$

and we have put $\sigma \xi^{d-1} = e_1 k_B T$ with e_1 of the order unity. Let us introduce four quantities:

$$Y_1 = D/P^{(s)} = C_3 \tilde{L} \tilde{t}^{-1} \quad [R_1 = (C_2/C_3) Z \tilde{L}] \ , \tag{4.9a}$$

$$Y_2 = D/P^{(e)} = C_4 \tilde{L}^{d} \tilde{t}^{-1} \quad [R_2 = (C_2/C_4) Z \tilde{L}^{2-d}] \ , \tag{4.9b}$$

$$Y_3 = I/P^{(s)} = C_2 C_3 Z \tilde{L}^{3} \tilde{t}^{-2} \quad [R_3 = \sqrt{R_1}] \ , \tag{4.9c}$$

$$Y_4 = I/P^{(e)} = C_2 C_4 Z \tilde{L}^{d+2} \tilde{t}^{-2} \quad [R_4 = \sqrt{R_2}] \ , \tag{4.9d}$$

where $C_3 = 1/(2a_1 e_1)$ and $C_4 = 1/(2a_0)$ are of the order unity, and we note that $C_3 = C_1 C_4$. R_i $(i = 1,2,3,4)$ are the Reynolds numbers in the case of $Y_i = 1$. The exponents a's in regions I, II, III, and IV are given by setting $Y_1 = 1$, $Y_2 = 1$, $Y_3 = 1$, and $Y_4 = 1$, respectively.

It is considered that the surface tension becomes effective for producing internal flows among droplets when droplets are percolated.[4] It is therefore considered that the surface tension is effectual for volume fractions larger than the percolation threshold v_p. However, even for volume fractions smaller than the percolation threshold v_p, a droplet always has other ones in its neighborhood with a certain probability. Thus, the effect of the surface tension cannot be neglected for off-critical concentration also. This may shift the exponent h in (1.6) and therefore the actual observed exponent a to larger ones (or to smaller ones). Such a shift in the exponent a may be explained as an anomalous elongation of the time-dependent crossover regime ("anomalous" also including "infinite").[12] Very recently Ohyama *et al.*[10] have reported the shift in the exponent a for a binary polymer mixture. Although they used a real-time and a real-length scale to analyze the experimental data, instead of the reduced ones, this yields no problem to examine the volume fraction dependence of the exponent a. Except for the temperature dependence, the observed result qualitatively agrees with a theoretical prediction.[12] The experimental data suggest that the diffusion constant D_T does not have a simple temperature dependence. The data also seem to suggest that the growth rate becomes slower as the temperature becomes lower far from the critical temperature. This is similar to certain model systems with large degeneracies.[9,12]

In this paper we temporarily neglect such an effect of "intermittent" growths of droplets.[12] Let v be the volume fraction of minority phase $(0 < v < 0.5)$. Then the percolation threshold may be roughly evaluated as $v_p = 2/z$, where z is the number of the nearest-neighbor droplets when the droplets are close-packed ($z = 6$ and 12 for $d = 2$ and 3, respectively) (see the first reference of Ref. 12).

Let us use the same estimations of the coefficients C's as Siggia.[4] Namely, we set (for $d = 3$)

$$C_3 \approx 3.0, \quad C_4 \approx 1/(12v) \quad (\text{and thus } C_1 \approx 36v) \ . \tag{4.10}$$

A rough estimate also gives

$$C_2 \approx 1 \ . \tag{4.11}$$

Then let us consider the region I, for example. This region is shown by $Q < 1$ and $R < 1$. $Q < 1$ is equivalent to $\tilde{L} > 6v^{1/2}$. On the other hand, the growth law in this region is given by setting $Y_1 = 1$. Therefore, the condition $R < 1$ is replaced by $R_1 < 1$, which is $\tilde{L} > 3v^{-1}$. Near the critical point Z is small. Thus, the region I near the critical point is $6v^{1/2} < \tilde{L} < 3Z^{-1}$. The growth law in this region is $\tilde{L} = 0.3\tilde{t}$. In this way the region of the applications of four growth laws can be known. They are illustrated in Fig. 2. A few remarks should be added. For smaller volume fractions such as $v < v_p = \frac{1}{6}$, the regions I and III do not occur. The region of the small Reynolds number lies between the regions of the large Reynolds numbers, since one of regions of large Reynolds numbers belongs to the region of large-Q value and the other to the region of the small-Q value. The growth laws at low temperatures are essentially the same as those at high temperatures. In practice, however, the regions I and II becomes very narrowed since $Z \approx 1$, and therefore the regions I and II are not observed at low temperatures. Thus, for $v < v_p$ and far away from the critical point, only region IV is applicable. For $v > v_p$ far away from the critical region, the region III also becomes applicable. Since the Reynolds number in this region increases indefinitely, the system becomes turbulent (turbulent growth). In the case of A of Fig. 2, the region I has no reality, since the droplet sizes in this region must be unphysically small.

The ratio Z defined by (4.8) depends on the dimensionality d. The kinetic viscosity may be evaluated as $\nu \approx r_0^2 / t_0$, where r_0 and t_0 are of the order of the atomic distance and the collision time, respectively. The kinetic viscosity may also be evaluated as $r_0 (k_B T / m_0)^{1/2}$ with $m_0 = g_0 / n_0$ and $n_0 \approx 1/r_0^3$. We can thus evaluate the ratio Z as

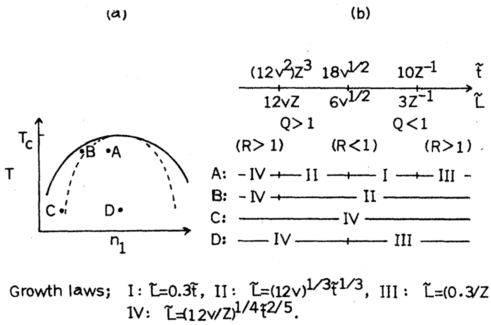


FIG. 2. Dominant droplet growth laws in three dimensions. "Dominant" means the neglect of the "intermittent effect" (Ref. 12) which may shift the growth rates and exhibit a wide crossover regime of the growth rates as actually observed.[10] In (a) the solid curve represents the two phase coexistence, and the dashed curve represents the percolation threshold. In (b) the upper horizontal line indicates the time axis. Lower four horizontal lines indicate the regions of the growth laws at corresponding times on the upper horizontal line. The symbols A, B, C, and D correspond to those in (a). Far away from the critical point (C and D) the regions I and II become very narrowed, and they are therefore omitted. In two dimensions the region corresponding to the region I disappears since $Z \approx 1$ for all temperatures. Z^3 on the time axis is replaced by Z^∞ and therefore the region II remains in two dimensions.

$$Z=C(r_0/\xi)^{d-2}\,, \tag{4.12}$$

with C being a constant of the order unity.

In two dimensions the region I disappears since Z is of the order unity independent of the temperature ($Z^{1/(d-2)}$ is the same as that in three dimensions). Except for the disappearance of the region I, the global behavior of the growth law in two dimensions is the same as that in three dimensions. In two dimensions we may set $v_p=\frac{1}{3}$.

V. DISCUSSION AND REMARKS

Experimental studies of phase separation of fluid mixtures are usually done in the very vicinity of the critical point. For instance, the reduced temperature $\epsilon=(T_c-T)/T_c$ is less than 10^{-4} for the isobuthyric acid + water mixture.[14] For such a region the ratio Z is smaller than 10^{-7} and the thermal correlation length ξ is larger than 10^{-5} cm and the kinetic viscosity is of the order $\nu \approx 0.1$. The Reynolds number (R_1) is less than unity for $\tilde{L}<10^7$. Therefore, reported experiments are those for small Reynolds numbers, corresponding to the growth laws in the regions I and II.[15]

There is no experiment for the real two-dimensional system. Therefore, a numerical simulation on argon[16] provides the only available data. Setting the average kinetic energy of an argon atom to be of the order 100 K, one may evaluate the collision time t_0 of an argon atom to be 1 ps. Therefore, the growth law corresponding to the regions II or IV is $\tilde{L}^2=\tilde{t}$, where $\tilde{L}$ is scaled by the diameter of the argon atom and $\tilde{t}$ is scaled by $t_0=1$ ps. The numerical simulation[16] was done at the density which seems to be smaller than the percolation threshold density $v_p=\frac{1}{3}$ (for $d=2$). Therefore, the growth law is for the region II or IV. The numerical simulation gives $\tilde{L}^2 \approx 0.6\tilde{t}$, which is consistent with the above theoretical prediction.

In connection with dynamic critical phenomena, the regions I and II correspond to the so-called critical regime, while the regions III and IV correspond to the so-called hydrodynamic regime.

The regions III and IV, where the inertia term becomes dominant, must be further examined, for instance, in connection with the turbulence, or with the generation of a turbulent state by stirring a phase-separating fluid.[17] This is, however, left for a future problem.

In the case of the region III (the turbulent growth $L \propto t^{2/3}$), the total kinetic energy which has been supplied, (3.1), is evaluated as

$$K_T=A-BL^{-1}\,, \tag{5.1}$$

where A and B are constant. Equation (5.1) means that the total kinetic energy supplied is equal to the released surface energy.

The growth law exponents a's in regions I and II are given by (1.4). For the regions I and II we may set $h=d-1$ and 0, respectively, together with $\theta=-\zeta=2$:

$$a=1/(d-h)\ \text{(for regions I and II)}\,. \tag{5.2}$$

On the other hand, for the regions III and IV, (1.4) does no longer hold. Instead, we have

$$a=2/(d+2-h)\ \text{(for regions III and IV)}\,. \tag{5.3}$$

Throughout this work we have not taken the gravitational effect into consideration. For fluid experiments the gravitational effect often becomes important for the late stage of the phase separation since two phases may have different mass densities. In the late stage of phase separation this may give a faster growth rate.[4] In this respect the problem of gravitational collapse of a star core after exhausting the nuclear fuel[18] may be a suitable application of the present theory. Since the star material is "falling," the star material does not feel the gravitational field. This problem is, however, outside the scope of this paper.

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APPENDIX A: FLUCTUATION-DISSIPATION RELATIONS

We assume that the fluid is in a thermal equilibrium state for a very short time interval of the order of the collision time of a fluid atom, say t_0. Then we replace the left-hand side of (2.1) by the local differentiation du/dt and we neglect the first term on the right-hand side of it. Then operating $\vec{\nabla}$ on both sides, and multiplying the both sides by $\vec{\nabla}\cdot\vec{u}(\vec{r}\,',t')$, we obtain

$$\begin{aligned}&\langle[\vec{k}\cdot\dot{\vec{u}}_{\vec{k}}(t)][\vec{k}\cdot\vec{u}_{-\vec{k}}(t)]\rangle\\&\quad-\int_{t-t_0}^{t}\langle[\vec{k}\cdot\dot{\vec{u}}_{\vec{k}}(t)][\vec{k}\cdot\vec{u}_{-\vec{k}}(t')]\rangle dt'\\&\qquad=-(\nu/c_1L^2)\langle[\vec{k}\cdot\vec{u}_{\vec{k}}][\vec{k}\cdot\vec{u}_{-\vec{k}}]\rangle\,,\end{aligned} \tag{A1}$$

where the overdot denotes the time derivative. The first term can be neglected due to the local equilibrium assumption. Thus, we obtain

$$(\nu/c_1L^2)(k_BT/g_0)k^2 = \int_{-\infty}^{t} \langle [\vec{k}\cdot\vec{f}^{(u)}_{\vec{k}}(t)][\vec{k}\cdot\vec{f}^{(u)}_{-\vec{k}}(t')] \rangle \; . \qquad (A2)$$

Here we have put

$$\langle [\vec{k}\cdot\vec{u}_{\vec{k}}][\vec{k}\cdot\vec{u}_{-\vec{k}}] \rangle = (k_BT/g_0)k^2$$

(equipartition law). The equality (A2) represents the fluctuation-dissipation theorem. With the use of this fluctuation-dissipation relation we can justify (2.10).

APPENDIX B: THE THERMAL DIFFUSION CONSTANT

Using the relations

$$\frac{d}{dt}n_k = -M_Tk^2\mu_k \; , \qquad (B1)$$

$$\mu_k = k_BTn_k / \langle n_kn_{-k} \rangle = k_BTn_k / (c_2n^2L^d) \; , \qquad (B2)$$

and setting the thermal mobility M_T as M for $L = \xi$ in (2.6), we obtain the diffusion constant (4.6).

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