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Crossover, locking-in, and intermittency of droplet growth rates in phase separation

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Nonuniversal behavior in the dynamics of phase separation is discussed. An equation which exhibits a nonuniversal growth rate in a long-time limit is derived relying on the dynamic-scaling assumption. Two contradictory behaviors, a crossover to larger growth rate and a formation of a locked-in structure, are shown to be described by this equation in two limiting cases. The intermittent region lies between regions with these two contradictory behaviors. Two types of intermittent-growth-rate exponents are obtained. One, a_{I1}, is valid at high temperatures, while the other, a_{I2}, is valid at low temperatures. These two exponents are, respectively, $a_{I1}=w_1a_1+w_2a_2$, and $a_{I2}=(w_1/a_1+w_2/a_2)^{-1}$. Here w_i $(i=1,2)$ are the probabilities of finding configuration associated, respectively, with the growth rates $R\propto t^{a_1}$ and $R\propto t^{a_2}$. a_1 is the largest exponent due to the curvature-driven force and a_2 is the next largest exponent; $a_2=0$ at zero temperature. Thus, for certain values of system parameters, the intermittent exponent a_I varies from 0 to a_{I1} as the temperature is increased. Several aspects of the growth rates in parameter space are predicted. They are consistent with numerical simulations and fluid mixtures.

I. INTRODUCTION

A phase separation in a quenched thermodynamically unstable system starts with the formation of small droplets and then the droplet coarsening follows.[1,2] When the average droplet radius R exceeds a certain value, say the thermal correlation length ξ, several aspects of the dynamics appear. These universal behaviors have been the subject of predictions and justifications of a universal law called a "dynamic-scaling law for the first-order phase transition."[3–7] An average quantity G as a function of wave number k and time t has a scaling form:

$$G_k(t)=[R(t)]^y\tilde{G}(kR(t)) , \tag{1.1}$$

where y is a constant. In order that the kinetic equation is consistent with the scaling assumption (1.1), the length scale R (droplet radius) must satisfy a simple power law for the growth rate,

$$R(t)\propto t^a , \tag{1.2}$$

with a constant exponent a. This scaling law has also been studied by many authors from theoretical[8] and experimental[9] viewpoints and now seems to be well established.[10] The largest growth rate is usually expected to dominate the final state. To show this we may write down, in a simple way, an equation of motion for droplet growth in which two different elementary processes are accounted for:

$$\frac{d}{dt}R=c_1R^{1-1/a_1}+c_2R^{1-1/a_2},\quad a_1>a_2 . \tag{1.3}$$

Such an equation may be obtained by deriving an equation of motion for structure function $S_k(t)$ and then using the scaling assumption:

$$S_k(t)=[R(t)]^d\tilde{S}(kR(t)) , \tag{1.4}$$

where d is the dimensionality. To derive (1.3) it is assumed that the scaling function $\tilde{S}(x)$ is independent of time t. This assumption is not, however, rigorous if more than two mechanisms are involved. Equation (1.3) is qualitatively correct and sufficient for the present purpose. In the large-R limit the second term on the right-hand side of (1.3) can be neglected and thus we observe that $R\propto t^{a_1}$. This means that the largest growth rate t^{a_1} always dominates the late stage of phase separation. The largest exponent a_1 is called a universal one. For relaxational systems the universal exponent is $\frac{1}{2}$ for nonconserved order parameters,[11] and $\frac{1}{3}$ for conserved order parameters.[12] For fluid mixtures the largest exponent is 1.[13] All these exponents can be derived through simple physical considerations on the curvatures of droplet surfaces.

Recent numerical simulations[14,15] on highly degenerate systems cast some doubt on the universal growth of droplets. The droplet growth exponent in a highly degenerate system is certainly smaller than a universal exponent. At low temperatures we also observe a "locking-in" phenomena, i.e., a system has a locked-in structure and droplets grow very slowly.[14–16] Such a locking-in can also be observed in a real system.[17]

Real systems have complicated structures. The locking-in in such systems might be attributed to their complicated structures. Therefore, even if the locking-in in such systems contradicts with a simplified evolution equation (1.3), no problem seems to arise. However, the numerical simulation[14] for a system with a simple lattice structure also exhibits a nonuniversal growth rate of droplets. The only characteristic of this system is that it has high degeneracies.

Traditionally, the occurrence of a locked-in structure is attributed to the formation of a droplet structure for which a curvature-driven force becomes ineffective.[11,18] For instance, a close-packed structure may render the curvature-driven force ineffective. Such a geometrically simple structure of a droplet, however, could not be possible. Thus, if we again adopt (1.3), then the locking-in becomes impossible in a long-time limit. In order to avoid such a difficulty, we have, in a series of papers,[19,20] proposed an intermittent process of droplet growth. In this process various growth mechanisms occur intermittently. Taking an ensemble average or a time average of $\ln R_j$ (R_j denotes the local droplet radius in the jth subsystem), we obtained a growth rate with a nonuniversal exponent smaller than the universal one, a_1. A qualitative agreement between our prediction and both numerical simulations and a fluid mixture experiment has been found.

Nevertheless, the previous derivation of the intermittent growth rate should be reexamined. The obtained theoretical intermittent growth exponent is independent of temperature. This contradicts numbers of numerical simulations and laboratory experiments. Such an insufficiency might come from two simplifications in the previous discussion. One of these simplifications is that we have neglected the effect of the lattice structure. At low temperatures the domain shape strongly depends on the lattice structure. The domain shape is crucial to the intermittent growth rate at low temperatures. The other simplification will be discussed in detail in this paper. This is related to the dynamic-scaling assumption for the switching mechanisms of droplet growth rates. The intermittent growth rate discussed in the previous papers[19,20] is valid only at high temperatures.

The purpose of this paper is to present the first step in a fundamental discussion of the intermittent growth law in the whole range of temperature. Very recently we have, in a short paper, proposed a theory for a crossover from a large to smaller growth rate.[21] Such a process competes with the process described by (1.3). In this paper it will be discussed that the intermittency is a true state and the intermittent region lies in between regions of these two competing processes. Namely, the intermittent-growth-rate exponent a_I lies between a_1 and a_2, where a_2 is the exponent of the next-most-dominant growth rate.

So far, the dynamic scaling assumption for the first-order phase transition is restricted to the study of the self-similarity of droplet configuration. Only a simple law of droplet growth rate (1.2) has been discussed. In this paper we shall apply the dynamic-scaling assumption to more complicated dynamical processes. One may find that the so-called universal droplet growth rates are not always universal; rather, nonuniversal growth rates may occur. We first show that the dynamic-scaling assumption is consistent with the intermittency. We then present a physical consideration of the appearance of intermittency. The appearance of the intermittency resembles the appearance of the gas-liquid transition. Namely, at certain values of system parameters, i.e., of temperature, degeneracy, and dimensionality, the final growth rate becomes marginal. Near this marginal point the effective time scale is extremely lengthened. This effect ensures the appearance of the intermittency. Various properties of intermittent growth of droplets are then predicted. Comparisons with numerical simulations and a fluid mixture experiment seem to verify theoretical predictions.

The present theory is by no means a complete one based on the first principle or the basic kinetic equation, but is devoted to the first step of the general description of the dynamics of phase separation in parameter space spanned by temperature, degeneracy, and dimensionality. In Sec. II we shall formulate an intermittent droplet growth on the basis of the dynamic-scaling assumption. In Sec. III a prediction will be made for the entire behavior of the growth rates in parameter space. The intermittent-growth-rate exponent a_I will be calculated in the two limiting cases of high temperatures and of low temperatures. Section IV is devoted to discussion. Several interesting behaviors of numerical simulation and fluid mixture experiment are also analyzed.

II. FORMULATION OF INTERMITTENCY

Let us consider small subsystems with a linear dimension of the order of R (the average droplet radius). Let the number of phases be p. We assume, for simplicity, that the average densities (volume fractions) of all phases are the same. Let us consider the case of high temperatures. On an average, the droplet of each phase takes the same shape. Thus the average droplet shape would be a close-packed one. The smallest degeneracy p_c for which the close-packed structure is possible is predicted to be $d+1$.[11,18] If, however, the random distributions of phases are allowed, then this value would be replaced by $z+1$ (Ref. 19) (z is the number of nearest-neighbor droplets in the close-packed configuration). It was also shown[19,20] that even for $p \geq z+1$ the close-packed structure does not always appear. A loose-packed structure appears with a certain probability which depends on p, d, and probably on temperature T. Thus the droplet grows for a certain time interval by means of a curvature-driven force, while for another time interval it grows by means of the next-most-dominant force. The next dominant force at higher temperatures is a thermal force and at low temperatures it is an exponentially weak force for relaxational systems.[19] In fluid systems the next-most-dominant force may be a curvature-driven force which drives individual atoms.[20] The largest growth rate occurs when the curvature-driven force induces internal flow.[13] The local droplet growth rate is switched back and forth from small to large and also large to small. Strictly speaking, only the curvature-driven force is switched back and forth. However, if the curvature-driven force is effective, then the next-most-dominant force may be neglected.

Since the droplet growth rate is very small, it takes a long time until a local growth rate is switched from one to the other (notice that this depends on what values system parameters take). Let us assume that the local droplet radius in the jth subsystem, R_j, obeys the equation of motion

$$\frac{d}{dt}R_j = \alpha_i R_j^{1-1/a_i}, \quad i=1,2 \; . \tag{2.1}$$

Here a_1 gives the largest exponent, while a_2 gives the next-largest exponent. These exponents are summarized in the previous papers.[19,20] Here we have listed them in Table I. If the switching of growth mechanisms is done

TABLE I. Various growth rate exponents of elementary processes.

Systems	Largest exponent a_1	Next-largest exponent a_2
Relaxational, nonconserved order parameter	$\frac{1}{2}$ [a]	$1/(d+1)$ [b] 0 (for $T\approx 0$) [c]
Relaxational, conserved order parameter	$\frac{1}{3}$ [d]	$1/(d+2)$ [e] 0 (for $T\approx 0$) [c]
Binary fluid mixture	1 [f]	$1/d$ ($d\leq 3$) [g] $1/3$ ($d>3$) [h]

[a] Reference 11.
[b] Reference 19.
[c] References 18 and 19.
[d] Reference 12.
[e] References 3 and 4.
[f] Reference 13.
[g] References 3 and 22.
[h] Reference 20.

frequently enough, one may obtain the same equation as (1.3). In this case no special effect due to the switching can be expected. At low temperatures or for high degeneracies the curvature-driven force often becomes ineffective. Once the system comes to a state in which the curvature-driven force becomes ineffective, it may take a long time for the system to escape from the state. Therefore, the switching of the local growth mechanisms is done very slowly. The average growth rate can not be represented by a simple sum of R_j^{1-1/a_1} and R_j^{1-1/a_2}.

In order to formulate an intermittent growth law, we shall make a simplification saying that the average radius also obeys the following equation similar to (2.1):

$$\frac{d}{dt}R=f_iR^{1-1/a_i} . \tag{2.2}$$

Here, however, the switching of the growth mechanisms is done with a short time interval. Let us assume that the ith growth mechanism continues for a short time interval Δt_i ($\ll t$). Then the increment of the local droplet radius for this time interval is given by

$$\Delta R_i=f_iR^{1-1/a_i}\Delta t_i,\quad i=1,2 . \tag{2.3}$$

We now notice that the ratio $\Delta t_2/\Delta t_1$ is not necessarily independent of t or R. Each growth process has its own growth rate. Therefore, it takes its own time to attain a significant increment of the droplet radius. The growth rate depends on the average droplet radius. Owing to the dynamic-scaling assumption, the ratio $\Delta t_2/\Delta t_1$ is proportional to a power of R:

$$\frac{\Delta t_2}{\Delta t_1}=\frac{w_2}{w_1}R^{1/a_I-1/a_1},\quad w_1+w_2=1 \tag{2.4}$$

where a_I is a constant whose meaning will become clear later. From (2.4) and (2.3)

$$\frac{\Delta R_2}{\Delta R_1}=\frac{f_2w_2}{f_1w_1}R^{1/a_I-1/a_2} . \tag{2.5}$$

Thus the average growth rate of R obeys the equation

$$\frac{dR}{dt}=\frac{\Delta R_1+\Delta R_2}{\Delta t_1+\Delta t_2}=f_1R^{1-1/a_1}\frac{1+(f_2w_2/f_1w_1)R^{1/a_I-1/a_2}}{1+(w_2/w_1)R^{1/a_I-1/a_1}} . \tag{2.6}$$

Let us assume that

$$a_2\leq a_I\leq a_1 . \tag{2.7}$$

Then, for large R, (2.6) reduces to

$$\frac{dR}{dt}=\frac{w_1f_1}{w_2}R^{1-1/a_I} . \tag{2.8}$$

This gives a nonuniversal growth rate $R\propto t^{a_I}$. If $a_I=a_1$, then (2.6) gives

$$\frac{dR}{dt}=w_1f_1R^{1-1/a_1}+w_2f_2R^{1-1/a_2},\quad a_I=a_1 \tag{2.9}$$

which was also given by Kawasaki.[23] If $a_I=a_2$, then (2.6) gives

$$\left[\frac{dR}{dt}\right]^{-1}=(w_1f_1+w_2f_2)^{-1}\times[w_1f_1(f_1R^{1-1/a_1})^{-1}+w_2f_2(f_2R^{1-1/a_2})^{-1}],\quad a_I=a_2 . \tag{2.10}$$

Equation (2.9) exhibits a crossover from a small growth rate $\propto t^{a_2}$ to a larger one $\propto t^{a_1}$. This is essentially equivalent to (1.3). On the other hand, (2.10) represents a crossover from a large growth rate to a smaller growth rate. Thus (2.10) exhibits a locking-in at low temperatures where $a_2=0$.

We now mention the physical meaning of these equations. As noted in Sec. I, if two processes with growth-rate exponents a_1 and a_2 occur in a parallel way then the equation can be given by (2.9). This means that the ratio $\Delta t_2/\Delta t_1$ is independent of R or t. The switching of growth mechanisms is done independent of the incrementation of droplet radius R. This means that the various configurations giving different growth rates are mixed up. Such a situation occurs at high temperatures or for low degeneracies. w_1 and w_2 are the weight functions and thus w_1 is the probability of finding a configuration giving the largest growth rate.

At low temperatures and for high degeneracies, the droplet configuration would be simple. If a system accidentally comes to a state where the curvature-driven force is ineffective, then the droplet must grow without the curvature-driven force. In order for the system to escape from the stationary state, local droplet radius R_j must be increased by an amount of the order of R without the curvature-driven force. Thus at low temperatures the switching between the growth mechanisms occurs with each constant (relative) increment of the local droplet radius. Then the duration times Δt_i $(i=1,2)$ are given by

$$\Delta t_i = (f_i R^{1-1/a_i})^{-1} \Delta R^{(i)} \, ,$$

$$\Delta R^{(i)} = w_i f_i \Delta R / (w_1 f_1 + w_2 f_2) \qquad (2.11)$$

with a constant ratio $\Delta R^{(2)}/\Delta R^{(1)}$. The time rate of change of R is then given by $dR/dt \approx \Delta R/(\Delta t_1 + \Delta t_2)$, which gives the equation (2.10). Thus (2.10) describes droplet growth in a system with high degeneracy at low temperatures.

At intermediate temperatures or for intermediate degeneracies the above two switching processes may mix together, or, the final state is equally dominated by the next-largest growth rate and by the largest growth rate. This fluctuation at the intermediate temperatures or the intermediate degeneracies would cause the intermittency. Owing to the dynamic-scaling assumption, the only plausible way of mixing together is given by (2.4), which gives an intermittent growth law as seen above.

III. PHYSICAL BACKGROUND OF INTERMITTENCY

Let us focus our attention on local droplet growths. The local length scale R_j does not increase as smoothly as the average droplet radius R. At high temperatures and for low degeneracies, switchings of local growth rates occur frequently, since the droplet configuration is so complicated. Several kinds of configurations are mixed up. In such a case the equation obeyed by the local length scale has the same form as Eq. (2.9). As temperature is lowered or the degeneracy is increased, the droplet configuration becomes less complicated. Time intervals between the switchings of local growth rates are then lengthened. Then the effective time scale becomes longer and the intermittent effect can be enhanced. In other words, a crossover regime can be lengthened. This is the reason why the intermittent growth law begins to dominate as the temperature is lowered or as the degeneracy is increased (see Fig. 1). Next let us consider the low-temperature limit or the high-degeneracy limit. We may consider that in such a limit the switching between the growth rates should be done with each constant relative increment of local droplet radius. Then we may obtain the same form for the local droplet radius R_j as given in Eq. (2.10). This equation shows a crossover from a large growth rate to a small growth rate. At low temperature this exhibits a locking-in, since $a_2=0$ at $T=0$.[19] At intermediate temperatures or for intermediate degeneracies the final growth rate is marginal: It is dominated by a large growth rate $\propto t^{a_1}$ as well as a small growth rate $\propto t^{a_2}$. Thus the temporal range of intermittent growth becomes very large. The intermittency thus has a marginal region. Let T_m and p_m be the temperature and the degeneracy for which the final growth rate cannot be determined. Then these values may be called "marginal values." T_m would be a function of p_m. Namely, as p_m increases, T_m also increases. The phase diagram of intermittency is thus qualitatively symmetric with respect to the line $T \propto p$ (see Fig. 2). If the transition to the intermittency is of first order, then T_m and p_m span a parameter space (see Fig. 2).

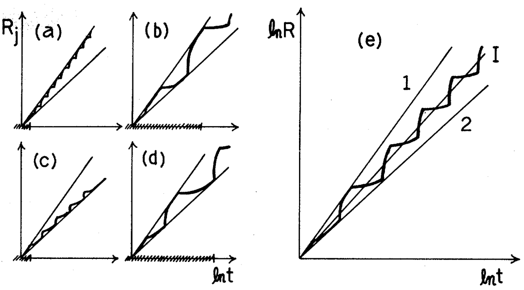

FIG. 1. Appearances of intermittency (schematic). (a) Switchings of local growth rates at high temperatures. Switchings are done in each constant relative time interval $(\Delta \ln t)_1$ or $(\Delta \ln t)_2$. A shaded region is a crossover region or an intermittent region. (b) Same as (a), but relative time intervals are lengthened, since the temperature is lower than in (a). Intermittent-growth-rate exponent a_I is $w_1 a_1 + w_2 a_2$. (c) Switchings of local growth rates at low temperatures. Switchings are done with each constant relative increment $(\Delta \ln R_j)_1$ or $(\Delta \ln R_j)_2$. (d) Same as (c), but the relative increments of local droplet radius are lengthened. Intermittent-growth-rate exponent a_I is $(w_1/a_1 + w_2/a_2)^{-1}$. (e) Switchings of average growth rate in a fully intermittent region. Here from (2.5) $(\Delta \ln R)_2/(\Delta \ln R)_1 \to 0$ as $R \to 0$.

As we have considered, the intermittent regime would emerge as a result of an elongation of a crossover regime. Thus the intermittent-growth-rate exponent a_I can be approximately given by the effective exponent at the crossover regime. If we assume that the growth mechanisms in the local droplet radius R_j are switched at considerably large time intervals, we can neglect the transient effect due to the switchings; then we have, by solving Eq. (2.1),

$$R_j = \left[\frac{\alpha_i}{a_i} t \right]^{a_i} \equiv R_j^{(i)}, \quad i=1,2 \, . \qquad (3.1)$$

It is assumed that the temporal region under consideration

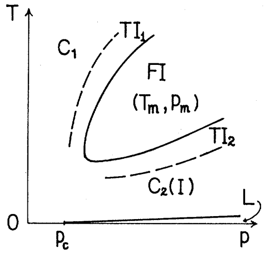

FIG. 2. Phase diagram for intermittency. C_1, region of crossover from small to larger growth rate. $C_2(I)$, region of crossover from large to smaller growth rate. This region is seemingly the same as an intermittent region, since the exponent a_2 in this region is temperature dependent. TI$_1$, temporary intermittent region for finite quenching time t, in a long-time limit the growth rate reduces to a universal one $R \propto t^{a_1}$. TI$_2$, temporary intermittent region, in a long-time limit the growth rate reduces to $R \propto t^{a_2}$. L, locking-in regime. Here p_c is the largest degeneracy for which the curvature-driven force is always effective.

is given by $R_j^{(1)} \approx R_j^{(2)}$. By averaging local growth rate equations, we have

$$\frac{d}{dt}R = \frac{1}{N}\sum_{j=1}^{N}\frac{dR_j}{dt} = \frac{a_1}{Nt}\sum{}^{(1)}R_j^{(1)} + \frac{a_2}{Nt}\sum{}^{(2)}R_j^{(2)} , \tag{3.2}$$

where N is the number of subsystems and $\sum^{(i)}$ denotes the summation over all subsystems in which the ith growth mechanisms $(i=1,2)$ dominate. We set

$$\frac{1}{N}\sum{}^{(i)}R_j = w_i R , \tag{3.3}$$

where w_i are the probabilities of finding subsystems with the ith growth mechanisms. For relaxational systems w_1 is the probability of finding a loose-packed structure and w_2 a close-packed structure.[19,20] From (3.2) and (3.3) we find that

$$\frac{d}{dt}R = \left[\frac{R}{t}\right](w_1 a_1 + w_2 a_2) . \tag{3.4}$$

Solving (3.3) we find the intermittent exponent

$$a_I = a_{I1} \equiv w_1 a_1 + w_2 a_2 = w_1(a_1 - a_2) + a_2 . \tag{3.5}$$

This exponent was previously obtained using various methods.[19,20] This exponent, a_{I1}, is valid at high temperatures or for low degeneracies, since (3.2) demands a constant ratio of time intervals between switchings.

Next we shall give another intermittent-growth-rate exponent a_{I2}, which may be suitable to the case of low temperatures and high degeneracies. We consider the elongation of the crossover regime at low temperatures. Let us assume that the switching between the growth mechanisms occurs with each constant relative increment of local droplet radius. We also neglect the transient effect due to the switchings of local growth mechanisms. Then the relative time interval for attaining a relative increment of local droplet radius in this process is

$$[\Delta \ln t(R_j)]_i = \frac{1}{a_i}(\Delta \ln R_j)_i , \quad i=1,2 . \tag{3.6}$$

Thus, setting

$$(\Delta \ln R_j)_i = w_i \, \Delta \ln R_j , \tag{3.7}$$

we have

$$\begin{aligned}\Delta \ln t(R_j) &= [\Delta \ln t(R_j)]_1 + [\Delta \ln t(R_j)]_2 \\ &= \left[\frac{w_1}{a_1} + \frac{w_2}{a_2}\right]\Delta \ln R_j .\end{aligned} \tag{3.8}$$

We then obtain another intermittent-growth-rate exponent:[21]

$$a_I = a_{I2} = \left[\frac{w_1}{a_1} + \frac{w_2}{a_2}\right]^{-1} , \tag{3.9}$$

which should be valid at low temperatures or for high degeneracies. Notice that $a_{I1} \geq a_{I2}$. The equality holds for $w_1 = 0,1$ or $a_1 = a_2$. If $a_2 = 0$, then $a_{I1} = w_1 a_1$, while $a_{I2} = 0$. The difference between these two intermittent-growth-rate exponents resembles that between the total electric conductivities of a parallel circuit (a_{I1}) and of a series circuit (a_{I2}).

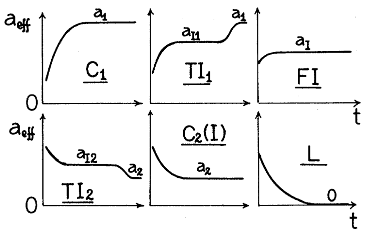

FIG. 3. Time dependence of the effective exponent $a_{\text{eff}} = d(\ln R)/d(\ln t)$. $C_1, C_2, \mathrm{TI}_1, \ldots$ represent the same parameter regions as in Fig. 2.

The present methods of obtaining intermittent-growth-rate exponents are linear ones. As noted in the above, the intermittent regime may be regarded as the anomalous elongation of the crossover regime. There are two possible types of the transitions from crossover regimes to intermittent regimes. If the transition is continuous, then the intermittency would continue for an infinitely long time only if the system parameters take on definite values. The whole behavior of the (effective) exponent $a_{\text{eff}}(t)$ $(R \propto t^{a_{\text{eff}}})$ is predicted in Fig. 3. Here the region $C_2(I)$ is the region where the growth rate always crosses over from a large growth rate $R \propto t^{a_1}$ to small growth rate $R \propto t^{a_2}$. a_2 may be switched from 0 at $T=0$ to nonzero values as temperature is increased and the final growth rate in this region depends on the temperature. Thus the final growth rate seemingly resembles the intermittent growth rate. The plateau value of a_{eff} gives an intermittent-growth-rate value a_I, such that a_I is zero at zero temperature. In particular, $a_I = a_{I2}$, and $a_{I2} = w_2 a_2 = 0$ at $T=0$ (see Ref. 19). As the temperature is increased, a_I also increases mainly due to the switching of a_2 from $a_2 = 0$ to $a_2 \neq 0$ (thermal contribution). As temperature is further increased, a_I reaches a constant value determined by a_{I1}. The whole behavior of a_I is illustrated in Fig. 4. In Fig. 5 we also illustrate the exponent a_I as a function of degeneracy p and the dimensionality d. In previous papers[19,20] we have found that such a dependence of the intermittent-growth-rate exponent a_I on degeneracy is ob-

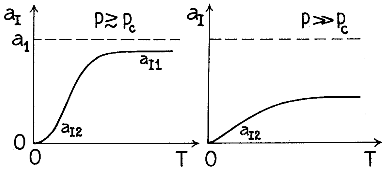

FIG. 4. Temperature dependence of intermittent-growth-rate exponent a_I. p_c represents the largest degeneracy for which the curvature-driven force is always effective, or the smallest degeneracy for which the intermittent growth rate with nonuniversal exponent occurs. The increase in a_I at low temperatures is mainly due to the increase in a_2 from 0 at $T=0$ to nonzero values at high temperatures.

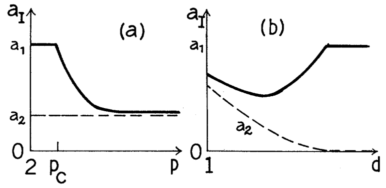

FIG. 5. (a) Intermittent-growth-rate exponent a_I as a function of degeneracy p. As temperature is lowered, both a_I in the high-p case and a_2 approach a zero value. (b) Intermittent-growth-rate exponent as a function of dimensionality. p_c is a function of d (see Ref. 19). This is the reason why a_I takes a universal value a_1 for large d. Since $a_1 = a_2$ for $d=1$ at high temperatures (see Table I), $a_I = a_1$ for $d=1$ at high temperatures.

served in numerical simulations and in a binary-fluid-mixture experiment. For a binary fluid mixture in three dimensions, the volume fraction of the minority phase corresponds to the reciprocal of the degeneracy, p^{-1}.[20]

IV. DISCUSSION

In this section we shall examine the predictions made in Sec. III. We shall compare these predictions with recent numerical simulations.[14,15] We have previously compared[19] the intermittency-growth-rate exponent a_{I1} given by (3.5) with numerical simulation of the p-state Potts model on a triangular lattice.[14] Improvements over the data of this simulation have recently been made. The agreement between the theoretical prediction and the simulation becomes poorer, though still satisfactory. In order to calculate w_1 we have employed a close-packed-cell model.[19,20] In this cell model, every cell is occupied by a single phase. The loose-packed structure appears when any neighboring cells are occupied by the same phase. When all phases have the same average density, random distributions of phases give the probability w_1 of finding a loose-packed structure:[19]

$$w_1 = \min\{zp^{-1}, 1\} . \tag{4.1}$$

Here z is the number of the nearest-neighbor cells, e.g., $z=6$ for $d=2$. In the case where the average densities of phases are not equal to each other, the probability may be given by[20]

$$w_1 = \min\left\{ z \sum_{i=1}^{p} n_i^2 ,\ 1 \right\} , \tag{4.2}$$

where n_i is the density of the ith phase. Here we have used units such that

$$\sum_{i=1}^{p} n_i = 1 . \tag{4.3}$$

The probability w_1 given by (4.1) or (4.2) might be suitable in the case of high temperatures. At low temperatures w_1 would strongly depend on the lattice structure. This might be the reason why the growth rate on a triangular lattice is different from that on a square lattice at low temperatures.[14] At high temperatures they are very similar. If w_1 is given by (4.1), then the curvature-driven force is always in effect for

$$p \le z . \tag{4.4}$$

On the other hand, there is another criterion different from (4.4) (Refs. 11 and 18):

$$p \le d . \tag{4.5}$$

This criterion [Eq. (4.5)] gives, however, the largest degeneracy for which the close-packed structure or effectively the same structure cannot be created. The smallest growth rate is the most dominant one at low temperatures (for certain system parameters). Therefore, the droplet configuration which gives the smallest growth rate is the most stable one at low temperatures near $T=0$. Thus (4.5) is suitable at low temperatures. On the contrary, (4.4) should be suitable at high temperatures. Thus w_1 must be a function of temperature.

In recent papers, Sadiq and Binder have reported[15] an observation on numerical simulation. The model which they used is a lattice-gas model on a square lattice with repulsion between nearest and next-nearest neighbors. The ground state has fourfold degeneracy ($p=4$). The order parameter (sublattice magnetization) is not conserved in this system. They performed simulation in two ways, i.e., by means of Glauber dynamics and by means of Kawasaki dynamics. In Glauber dynamics the magnetization of the Ising model is not conserved; in Kawasaki dynamics it is conserved. The ordering processes should be different in both cases. In the case of Kawasaki dynamics, the extension of the ordered domain is caused by the flow of particles in bulk. This corresponds to the dynamics of a system with conserved order parameter. Therefore the growth rate of the ordered domain in Kawasaki dynamics would be the same as in the case of the conserved order parameter. In the case of Glauber dynamics, the growth rate of the ordered domain is found to be (1.2) with $a=\frac{1}{2}$. On the other hand, the growth rate in the case of Kawasaki dynamics is not so simple. They examined effective growth rates of energy at several temperatures and at several quenching times in Kawasaki dynamics. We may consider the change in the energy as simply due to the change in the surface area. Their observations are as follows. (i) At low temperatures, the exponent y_{eff}, which may correspond to our exponent a, approaches zero with time, showing a locking-in. (ii) At higher temperatures (except in the vicinity of the critical point), the exponent y_{eff} increases from 0.27—0.28 to about 0.35, and then seems to stay at the final value ≈ 0.35. These values are, however, accompanied with large error bars. (iii) At intermediate temperatures the exponent y_{eff} seems to fluctuate between 0.2 and 0.3. The behavior of y_{eff} in region (i) can be explained if we assume that the criterion (4.5), i.e., $p \le 2$ is suitable. The thermal force cannot apply and therefore the smallest growth rate with $a_2=0$ is the most dominant. The dynamics may be described by (2.10) with $a_1=\frac{1}{3}$ and $a_2=0$. In region (ii), the criterion (4.4) is applicable. Since $p=4<6$, the dynamics is described by (2.9) with $a_1=\frac{1}{3}$ and $a_2=\frac{1}{5}$. the region (iii) must be an intermittent region where the intermittent-growth-rate exponent a_I seems to lie between $\frac{1}{3}$ and $\frac{1}{5}$. Their observations are the same as in Fig. 3. w_1 should depend on the temperature, however. Why is the intermittent effect not observed in their simulation in the case of Glauber dynamics? The reason might be as follows. There is an ambiguity in the threshold value p_c at which the intermittency effect appears, namely, $d+1 \le p_c \le z+1$ ($z=6$ for $d=2$) [see (4.4) and (4.5)]. p_c strongly depends on the cluster shape and cluster configuration. For Kawasaki and Glauber dynamics the cluster shapes and cluster configurations might be different. Thus the appearance of the intermittency in both cases may be different.

Similar behavior, as in Fig. 4, is seen in a numerical simulation done by Sahni *et al.*[14] The simulation was done using the Potts model with attractive force for $p=6$ and 36 on a square lattice with nonconserved order parameter. Due to high degeneracies both cases, $p=6$ and 36, exhibit locking-in at low temperatures near $T=0$. Otherwise these systems are intermittent with $a_I = a_{I2} = 0$ at $T=0$. The increase in a_I near $T=0$ is due to the switching of a_2 from $a_2=0$ to $\frac{1}{3}$. This switching should occur in a parallel way with certain probabilities $\tilde{w}_1$ and $\tilde{w}_2$. Thus, near $T=0$, one might set $a_2 = 0 \times \tilde{w}_1 + \frac{1}{3}\tilde{w}_2 = \frac{1}{3}\tilde{w}_2$. Assuming that there is an activation energy Δ for the process with $a_2 = \frac{1}{3}$ we may set

$$a_2 = \frac{1}{3} \frac{1+e^{\Delta/k_B T_c}}{1+e^{\Delta/k_B T}} , \tag{4.6}$$

where Δ is of the order of $k_B T_c$. Since this system exhibits the locking-in at $T=0$, the threshold value p_c at low temperature is smaller than $z=6$. For the sake of simplicity we neglect the temperature dependence of w_1. Instead of this simplification, we set $w_1 = \frac{5}{6}$ ($w_1 < 1$) for $p=6$. This ensures the locking-in at low temperatures and also gives the suitable value $a_{I1} = 0.47$ at high temperatures. For $p=36$, (4.1) gives $w_1 = \frac{1}{6}$. This gives $a_{I1} = 0.36$ at high temperatures, which is in good agreement with the experimental value. In Fig. 6 we compare theoretical intermittent exponents with numerical simulation. We note that the increase in the intermittent exponents near $T=0$ is due to the increase in a_2. The temperature dependence of w_1 is not important to this behavior of the intermittent exponent. Here we have chosen $\Delta = 0.4 k_B T_c$.

Sahni *et al.* also did the simulation using the Potts model on triangular lattice. A part of their results was examined in a previous paper.[19] No temperature dependence of the droplet growth rate exponent was observed on the triangular lattice.[14] On the other hand, the growth rate exponent on a square lattice strongly depends on temperature. This difference may be explained as follows. On a square lattice kinks of interface are absorbed by vertices. At low temperatures, therefore, the flexibility of the interface is lost on a square lattice.[14] On the other hand, there are many combinations between kinks and vertices on a triangular lattice. Some of the kinks can be absorbed by vertices, but other kinks remain without being absorbed.[14] Since there are many kinks in the initial disordered state, and most of the kinks would remain in a phase-separated state on a triangular lattice, the flexibility of the interface would be maintained even at low temperatures on a triangular lattice. For this reason the growth rate of the droplet on a triangular lattice does not exhibit temperature dependence, while it does on a square lattice. The flexibility of the interface on a square lattice depends on the density of the kinks, which are thermally generated with a probability such as $e^{-\Delta/k_B T}$. For a triangular lattice $\Delta = 0$ due to the "conservation" of kinks. Such a factor would appear in the probability w_1, though it is neglected in Fig. 6.

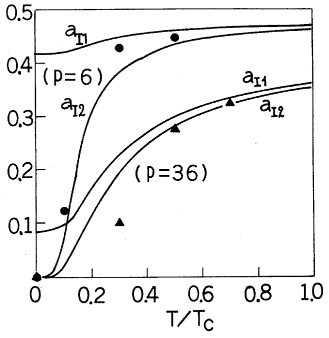

FIG. 6. Temperature dependences of a_{I1} and a_{I2} given by (3.5) and (3.9), respectively. Here it is assumed that $w_1 = \frac{5}{6}$ for $p=6$ instead of neglecting the temperature dependence of w_1. w_1 for $p=36$ is given by (4.1), i.e., $w_1 = \frac{1}{6}$. a_2 is given by (4.6) with $\Delta = 0.4 k_B T$. Experimental data are taken from Fig. 15 of Ref. 14(b) (Potts model with nonconserved order parameter on square lattice). Data should be compared with a_{I1} at high temperatures and with a_{I2} at low temperatures.

For binary fluid mixtures the same intermittent effect can be expected, since the largest growth rate due to the curvature-driven force, i.e., $R \propto t$, is possible only if internal flows are induced.[13] The internal flows are induced if the loose packing occurs for a minority phase. The probability of finding a loose packing of a minority phase is evaluated as[20]

$$w_1' = \min\{z n_2, 1\} , \tag{4.7}$$

where n_2 is the density of a minority phase and is equivalent to the volume fraction v. The intermittent-growth-rate exponent is given by

$$a_{I1} = w_1' + \tfrac{1}{3}(1 - w_1') \quad \text{for } d=3 \tag{4.8}$$

which is, at most, about 10% larger than a_{I2}, however. It has been shown[20] that this growth law agrees with experimental data at intermediate temperatures.[24] For lower temperatures, the growth rate of the experiment in the same range of volume fractions is given by $a \approx \frac{1}{3}$. This is, however, not unexpected. At lower temperatures the intermittent region may be narrower, and the final growth rate is $R \propto t^{1/3}$ (in a classical fluid the exponent $a_2 = 0$ is impossible owing to the appearance of the solid state).

V. REMARKS

There are two factors which cause the intermittency in the droplet growth. One is the existence of many competing growth mechanisms. The other is the existence of two different switching processes of these competing growth mechanisms. The phase diagram of the intermittency (Fig. 2) is the analog of the phase diagram of the gas-liquid transition (static). The fully intermittent region (FI) corresponds to the coexistence region. The occurrence of the intermittency resembles the occurrence of the condensation in the gas-liquid transition. This suggests the existence of intermittent critical phenomena and other phenomena found in the static phase transition.

We have given a discussion which is oversimplified in order to obtain the temperature dependence of a_{I2} at low temperatures [see (4.6)]. Such a discussion is only for a temporay purpose and should be replaced by a more microscopic discussion such as vertex dynamics,[14] which depends on the basic lattice structure. The temperature dependence of the intermittent-growth-rate exponent is, therefore, sensitive to the lattice structure.

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