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Domain growth with time-dependent front velocity in one dimension

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The kinetics of domain growth associated with a first-order phase transition is studied in a system with multiple-degenerate ordered states. Without assuming the constant growth rate of the nucleated domains, we derive a formula for the time evolution of the volume fraction of the growing domains in one dimension. Computer simulations show very good agreement with the theory. A rigorous relation for the two-point correlation function is also obtained.

I. INTRODUCTION

The kinetics of a first-order phase transition is governed by nucleation and growth of domains in the stable phase. Just below a first-order phase transition point the high-temperature disorder phase becomes metastable. Decay of the metastable state proceeds by nucleation of the low-temperature phases with the aid of thermal fluctuations. Domain growth after nucleation has been a subject of both theoretical and experimental studies for many years. Almost all the previous theories[1–5] are, however, restricted to the case of nondegenerate ordered phase and of the constant growth rate.

In a previous paper, Ohta *et al.*[5] have studied the kinetics of domain growth in systems with multiple-degenerate ground states and with a nonconserved order parameter. The system is characterized by a degeneracy parameter p that is equal to the number of degenerate but physically distinct stable low-temperature phases. In many experimental situations such as ferroelectric and structural transitions, the ordered phases are often degenerate.[6,7] This motivated them to deal with a statistical dynamics of degenerate domains.

Ohta *et al.*[5] have derived for arbitrary values of p the exact nonequilibrium scattering function of the newly forming domains nucleated from the metastable uniform state. This is an extension of the study by Sekimoto[3] where the case $p=1$ was formulated. These two theories give us not only the correlation functions but also, as a special case, the volume fraction of the ordered domains developed Kolmogorov[1] and Avrami[2] many years ago.

These theories are based on the assumption that the growth rate is constant and the domains are spherical. That is, the velocity of the domain front separating the domain and the surrounding metastable state is independent of time or the radius of domains. Although the constant growth rate enables us to formulate the theory of domain growth in a rigorous manner, it is a very restricted assumption in reality. The time evolution of the domain front is generally affected by such as the interfacial tension and the elastic field. For instance, expansion of a domain increases the domain front area and hence it is energetically unfavorable. Thus the front velocity decreases as the domain size increases. Furthermore, as is well known, the critical radius of nucleation becomes finite when the interfacial energy is taken into account. If one wishes to formulate this case, one has to regard that domains grow at infinite velocity up to the critical radius and then at a finite velocity. The theory of a constant growth rate cannot be applied even if the front velocity after nucleation is independent of time.[3] These two are the example where the domain front velocity is a decreasing function of its size. When the growing stable phase has a different lattice structure from the metastable matrix, the elastic strain around the domains affects the growing kinetics drastically.[8,9] It has been shown that, if domains are softer than the matrix, the domain growth is decelerated while if those are harder the front velocity tends to increase. The last case is an example that the front velocity is an increasing function of its size.

Because of the lack of theories for a time-dependent growth rate, one often encounters a difficulty in analyzing experimental data. That is, if the observed volume fraction does not fit into the Kolmogorov-Avarmi formula allowing the time dependence of the nucleation rate,[5] one may ascribe this to the time dependence of the growth rate. However, because the corresponding theory is not available, it is impossible to determine its explicit form and to clarify uniquely the underlying physics within the data.

In this paper we address the domain growth problem without assuming the constant growth rate. Our theory does not rely on any specific forms of the growth rate. What we impose is that the front velocity is a monotonous function of time. For simplicity, we restrict ourselves mainly to one dimension. Hence the argument concerning the origin of the time dependence of the growth rate mentioned above cannot be applied since the

interfacial energy does not play any decisive role of the kinetics in one dimension. Nevertheless, the present theory shows explicitly how the time evolution of the volume fraction is influenced by the nonconstant growth rate. Some of the properties are expected to hold qualitatively even in higher dimensions. In fact, it will be found that the theory for decreasing growth rate, which is more frequently observed experimentally than the increasing case, can be extended to higher dimensions.

Although other assumptions are the same as those in Ref. 5, we summarize here the model to make the paper self-contained. Nucleation obeys the Poisson distribution and the nucleation rate may depend on time. We do not consider any correlation between nucleation events. There is no interaction between the growing domains except for a direct collision. That is, when two domain fronts collide with each other, the interface disappears if two domains are the same ordered phase whereas it ceases to move if the domains belong to different phases. Thus the system for $p > 3$ eventually exhibits a cell pattern of the ordered domains. We do not consider the further coarsening of the cell structure.

In the next section, the volume fraction of the ordered domains is calculated by allowing the time dependence of the front velocity. We treat the cases of the increasing and the decreasing front velocities separately. Here we are concerned with a one-dimensional system. In order to verify the accuracy of the theory, we carry out, in Sec. III, Monte Carlo simulations. In Sec. IV, we derive an exact relation for the two-point correlation functions for arbitrary dimensions. This relation can extract the explicit p dependence of the correlation function of the ordered domains. The only assumption used in this derivation is the equivalence and the independence of p-ordered phases. The method to obtain the volume fraction described in Sec. II cannot be applied to higher dimensions especially when the front velocity increases with time. This is discussed in the concluding remarks given in the final section (Sec. V).

II. VOLUME FRACTION OF THE ORDERED DOMAINS

We consider the situation such that by a temperature quench the system at the high-temperature phase is brought into the metastable state below but near the first-order transition point and the nucleation of the low-temperature ordered phases is initiated. The domain growth is characterized by the the volume fraction and the spatial correlation function of the ordered domains. In this section we study the time evolution of the volume fraction.

As in Ref. 5 we assume that the nucleation event obeys the Poisson process. The nucleation rate which is the probability of nucleation per unit space-time volume is denoted by $I(t)$ at time t after quench. The system is translationally invariant and is extended infinitely in d dimensions.

Before entering the theory for the time-dependent growth rate, we briefly review the essential part of the theory in Ref. 5. In the study of domain growth, it is most convenient to consider the trajectory of the motion of a domain front in the $(d+1)$-dimensional space-time coordinate. Suppose that we are concerned with the state at position $\mathbf{r}$. In order to cover this point by one of the ordered domains at time t, a nucleation event has to occur at the surface of a cone whose apex is at the point $\mathbf{r}$ and t in the $(d+1)$-dimensional space. The cone defined in this way is called a casual cone.[5] An example of the causal cone for an increasing growth rate in one dimension will be given in Fig. 1 below. The volume $\Lambda^{(1)}(t)$ of the causal cone weighted by the nucleation rate $I(t)$ plays a central role in the theory. It is defined by

$$\Lambda^{(1)}(t)=\int_0^\infty d\tau I(\tau)V(t-\tau) , \tag{2.1}$$

where

$$V(t)(=0 \text{ for } t<0) \tag{2.2}$$

is the d-dimensional volume of the nucleated domain.

The volume fraction is obtained easily by using the causal cone when the domain front velocity is time independent. Let $\phi(t)$ be the volume fraction of the metastable domain at time t. The volume fraction $\psi(t)$ of the ordered domains is then given by

$$\psi(t)=1-\phi(t) . \tag{2.3}$$

It is noted that $\phi(t)$ is equal to the probability of covering a point by the metastable domain at time t. Now we calculate the probability that a point $\mathbf{r}$ is covered by one of the ordered domains for the first time in the infinitesimal interval between t and $t+dt$. This is given by $\phi(t)d\Lambda^{(1)}(t)$ since the probability that the point $\mathbf{r}$ is uncovered is equal to $\phi(t)$ and the probability of nucleation of one of the ordered domains in the shell between $\Lambda^{(1)}(t)$ and $\Lambda^{(1)}(t+dt)$ is given by $d\Lambda^{(1)}(t)>$ Integrating the above probability from 0 to t, we obtain[5]

$$\psi(t)=1-\exp[-\Lambda^{(1)}(t)] , \tag{2.4}$$

where we have used the fact that

$$\phi(t)=\exp[-\Lambda^{(1)}(t)] . \tag{2.5}$$

This is a consequence of the Poisson process of the nucleation events. A simpler derivation is to start with (2.5) and use (2.3). The volume fractions $\phi(t)$ and $\psi(t)$ are related to $\phi_0(t)$ and $\phi_1(t)$ defined in Ref. 5 as $\phi(t)=\phi_0(t)$ and $\psi(t)=p\phi_1(t)$.

Now we extend this method to the time-dependent domain growth. First we consider the case where the front velocity is an increasing function of time. Here we restrict ourselves to a one-dimensional system. A difficulty to generalize to higher dimensions will be described in Sec. V.

In one dimension, the domain area is given by

$$V(t)=2\int_0^t dt_1 v(t_1) , \tag{2.6}$$

where $v(t)$ is the front velocity and the time origin in (2.6) is the instance when the domain is nucleated. We evaluate the probability that the point $x=0$ in Fig. 1 is covered in the interval between t_1 and t_1+dt_1 for the first time by an ordered domain. The cone with its apex

at $t=t_1$ and $x=0$ is a causal cone. As mentioned above, domains which are nucleated only on the surface of the cone have the possibility to cover the point $t=t_1$ and $x=0$. Suppose that a nucleation occurred at point A in Fig. 1. Two thin lines (inverted cone) which meet each other at point A represent the trajectory of the domain fronts after nucleation. As is clearly seen from Fig. 1, this nucleated domain can cover $x=0$ only if there is no nucleation even in the hatched area. Thus the probability is given by

$$dt_1\phi(t_1)2\int_0^{t_1}dt_2 v(t_1-t_2)I(t_2)\exp[-f(t_1,t_2)]\equiv dt_1\phi(t_1)F(t_1)\ , \tag{2.7}$$

where $f(t_1,t_2)$ is the weighted volume of the hatched area:

$$f(t_1,t_2)=\int_{t_2}^{t_1}dt_3 I(t_3)\left[\int_{t_3-t_2}^{t_1-t_2}ds\, v(s) - \int_0^{t_1-t_3}ds\, v(s)\right]. \tag{2.8}$$

It is noted here that $\phi(t_1)$ in (2.7) is not given by (2.5) but an unknown quantity to be determined as below. We obtain $\psi(t)$ after integration over t_1 from 0 to t as

$$\psi(t)=\int_0^t dt_1\phi(t_1)F(t_1)\ . \tag{2.9}$$

Using the relation $\psi(t)=1-\phi(t)$ and the fact that $\phi(0)=1$, Eq. (2.9) can readily be solved as

$$\psi(t)=1-\exp\left[-2\int_0^t dt_1\int_0^{t_1}dt_2 v(t_1-t_2)I(t_2)\times\exp[-f(t_1,t_2)]\right]. \tag{2.10}$$

This is the formula of the volume fraction for an increasing domain front velocity. When $v(t)$ is independent of time, $f(t_1,t_2)$ vanishes so that (2.10) reduces the Komolgorov-Avrami formula (2.4).

Here a remark is now in order. In the derivation of (2.10) we have assumed implicitly that the point A in Fig. 1 belongs to the metastable states so that nucleation of the ordered phase is possible. One might doubt that this is not always justified. In fact, if there occur two successive nucleation events as in Fig. 2, the region m-n on the surface of the causal cone is covered by the ordered domain without covering the point $x=0$ at time t_1. However, since we have imposed the condition that there is no nucleation event in the hatched region in Fig. 1, such a process does not enter in the calculation of the probability (2.7) and it is legitimate to assume that the surface of the cone remains at the metastable phase.

Next we consider the case where $v(t)$ (>0) is a smoothly decreasing function of t. The causal cone takes the form shown in Fig. 3. Again we calculate the probability such that the point $x=0$ is covered by an ordered domain for the first time at $t=t_1$. The domain front nucleated at point A in Fig. 3 reaches to $x=0$ at $t=t_1$. However this process is not possible if there was a nucleation in the hatched region since the point A has been covered by the ordered domain and does not belong to the metastable state. Thus we have to take into account the probability of no nucleation in the hatched region, which is given by $\exp[-g(t_1,t_2)]$ where

$$g(t_1,t_2)=\int_0^{t_2}dt_3 I(t_3)\left[\int_0^{t_1-t_2}ds\, v(s)+\int_0^{t_2-t_3}ds\, v(s) - \int_0^{t_1-t_3}ds\, v(s)\right]. \tag{2.11}$$

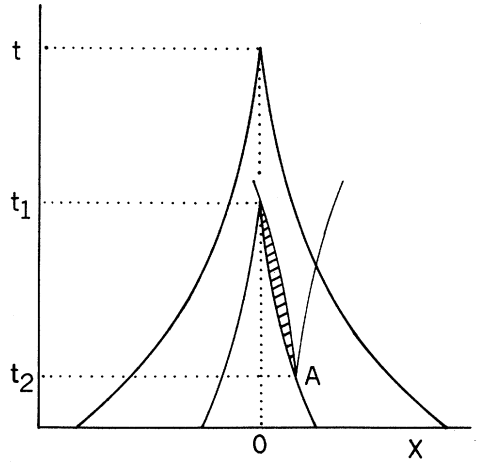

FIG. 1. The causal cones with the apex at $t=t$ and $t=t_1$ for the increasing growth rate.

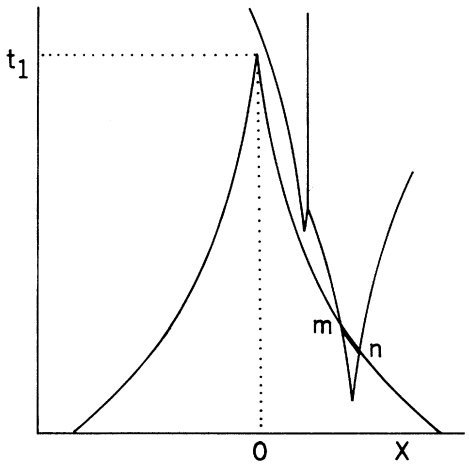

FIG. 2. The case where the region m-n is covered by an ordered domain.

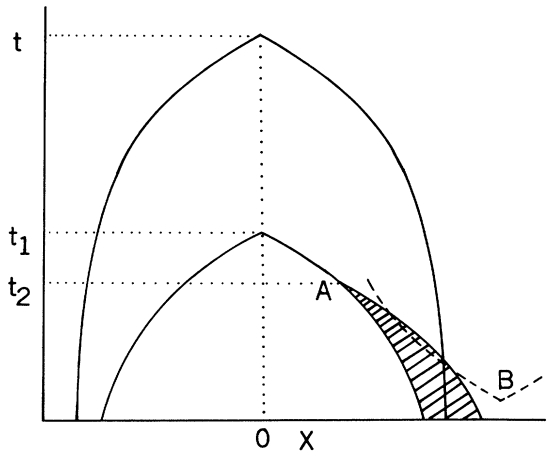

FIG. 3. The causal cones with the apex at $t=t$ and $t=t_1$ for the decreasing growth rate.

By the method similar to the derivation of (2.10) we obtain

$$\psi(t)=1-\exp\left[-2\int_0^t dt_1\int_0^{t_1} dt_2 v(t_1-t_2)I(t_2)\times\exp[-g(t_1,t_2)]\right] . \quad (2.12)$$

When $v(t)$ is constant, $g(t_1,t_2)$ vanishes so that the formula (2.12) agrees with (2.10) and the previous result (2.4).

Finally we consider the domain growth with a finite critical radius of nucleation. The front velocity is written as

$$v(t)=x_c\delta(t)+u(t) , \quad (2.13)$$

where $2x_c$ is the critical width of domains and $u(t)$ is the growth rate after nucleation. Equation (2.13) cannot be directly applied to Eq. (2.12) since it is valid only for a smooth function $v(t)$. However a similar consideration leads us to

$$\psi(t)=1-\exp\left[-\frac{2v_0}{x_c}t+2\frac{v_0}{x_c^2 I_0}(1-e^{-tI_0x_c})-2E(x_cI_0t)\right] . \quad (2.14)$$

Here, for simplicity, we have put $u(t)=v_0=$const and $I(t)=I_0=$const. The function $E(x)$ is defined by

$$E(x)=\int_0^x ds\,\frac{1}{s}[1-\exp(-s)] . \quad (2.15)$$

In the limit $x_c\to 0$, (2.14) agrees with the known result

$$\psi(t)=1-\exp(-v_0I_0t^2) . \quad (2.16)$$

This is obtained from (2.4). One of the features of (2.14) is that $\phi(t)$ in the limits $t\to\infty$ exhibits the exponential decay with the power-law correction, i.e., $\phi(t)=(x_cI_0t)^{-2}\exp[-(2v_0/x_c)t]$. This is a clear distinction from (2.16).

The formula derived in this section is not rigorous. For instance, we have put the condition of no nucleation in the hatched region in Fig. 3 for the decreasing case. This assumes that the hatched region belongs to the metastable phase. However, if there is a nucleation shown by B, a part of the hatched region is covered by the ordered domain so that the region of the metastable phase becomes smaller. It seems impossible to take rigorously this hierarchical process into consideration.

III. COMPUTER SIMULATIONS

Here we present the results of computer simulations in one dimension. The system size (lattice points) L is chosen as $L=1560$. However we have used only the data in the middle of the system with the size $L/2$ to avoid the

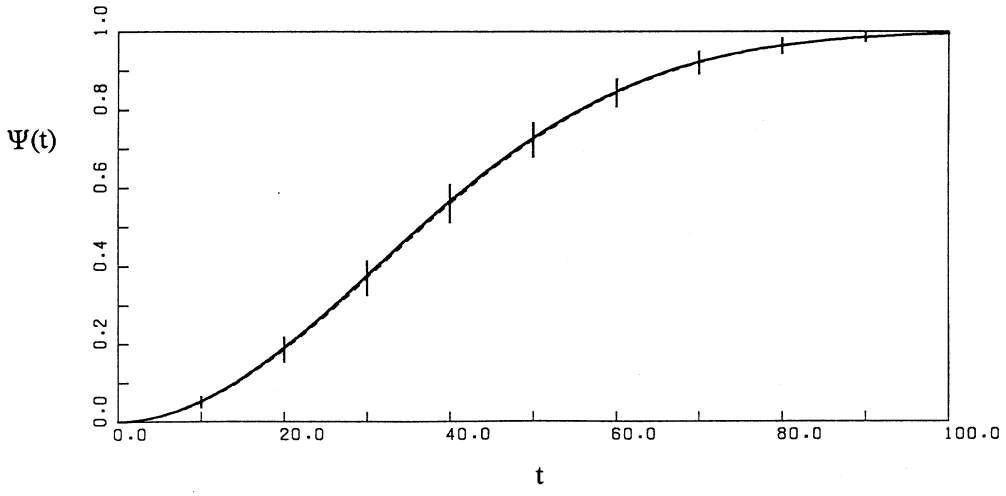

FIG. 4. The time dependence of the volume fraction for the constant growth rate. The vertical axis is $\Psi(t)$ as a function of time t in the horizontal axis.

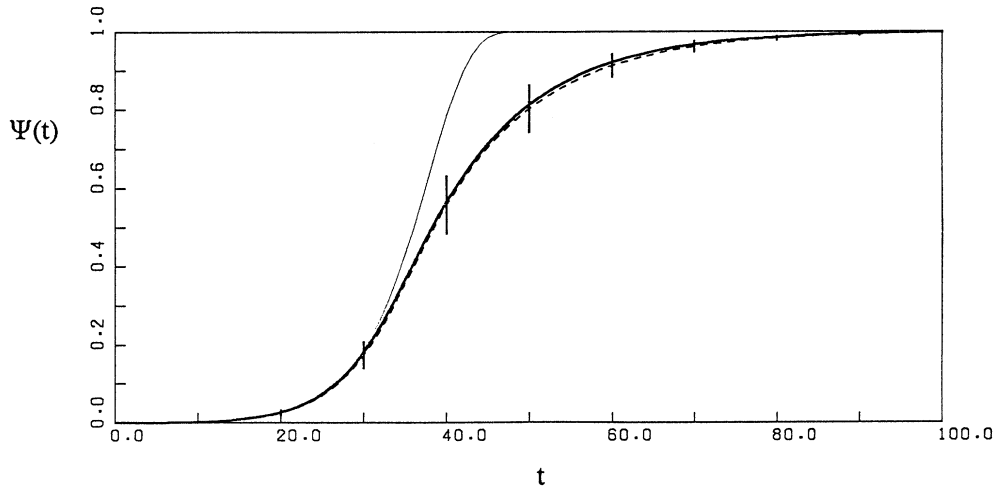

FIG. 5. The time dependence of the volume fraction for the increasing growth rate. The meanings of the axes are the same as those in Fig. 4.

boundary effect. In order to see the consequence only of the time-dependent growth rate, we put the nucleation rate $I(t)$ to be constant, $I(t)=I_0=0.8/L$. The front velocity is chosen as $v(t)=ab\exp(\pm bt)$ with $a=0.1$ and $b=0.2$ for the increasing $(+)$ and with $a=100$ and $b=0.2$ for the decreasing $(-)$ cases. The reason as to why we use the exponential growth rate is simply because it enables us to extract the effects of the time dependence most pronouncedly. The critical radius of nucleation is put to be zero.

The domain growth is simulated as follows. Initially the system is in the metastable disorder state. All the lattice points are assigned by the number "0." We generate a uniform random number c which is in the interval between 0 and 1 and specifies the possible nucleation point $x=cL$. Then, we again generate a random number. If this number is smaller than 0.8, we regard that nucleation occurs at that point and assign the number 1, while if not, the state of the point remains unchanged. Since we are concerned with the volume fraction, it is unnecessary to consider the degeneracy of the ordered phase. The nucleated domain is then expanded in the magnitude $v(t)\Delta t$ where the increment of time is chosen as $\Delta t=0.005$. Then we return to the first step. If the point specified by $x=cL$ has already belonged to "1," we discard this point and simply expand the domains.

In order to check the accuracy of simulations we first performed simulations for the constant velocity $v(t)=1$. Other conditions are the same as described above. Figure 4 shows the obtained volume fraction $\psi(t)$ as a function of time. The broken line is the average of 100 independent runs. The vertical bars indicate the typical scatter of the data. The full line is the rigorous result given by Eq. (2.4) or more explicitly by (2.16). Note that two lines are almost indistinguishable. This confirms the accuracy of simulations.

The result for the increasing front velocity is shown in Fig. 5. The broken line is the average of 100 runs with the deviation indicated by the vertical bars. The thick line is our theoretical result given by Eq. (2.10), which is in very good agreement with the simulations. For comparison, the Komogorov-Avrami formula (2.4) substituted by the time-dependent velocity is also shown by the thin line. The reason as to why the approximate result (thin line) gives the rapid increase of $\psi(t)$ compared to our theoretical result and simulations is obvious. Suppose that there are two growing domains as in Fig. 6. One (A) is nucleated earlier than the other (B). If the domain (B) were absent, the domain (A) would grow as shown by the dotted line. However when the second nucleation occurs, the left front of (A) is blocked. Thus the covering of the system is effectively delayed in the case of the increasing velocity. This process is not incorporated to the approximate theory given by the thin line.

Figure 7 displays the volume fraction for the decreas-

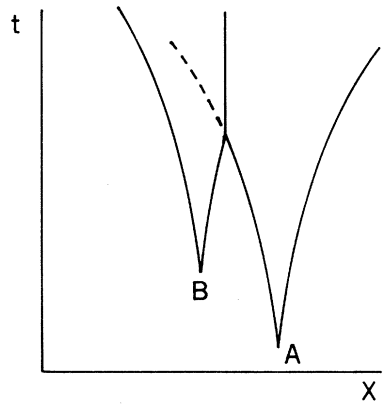

FIG. 6. A block of A domain by B domain. The dotted line indicates the trajectory of the left front of A when it was not blocked.

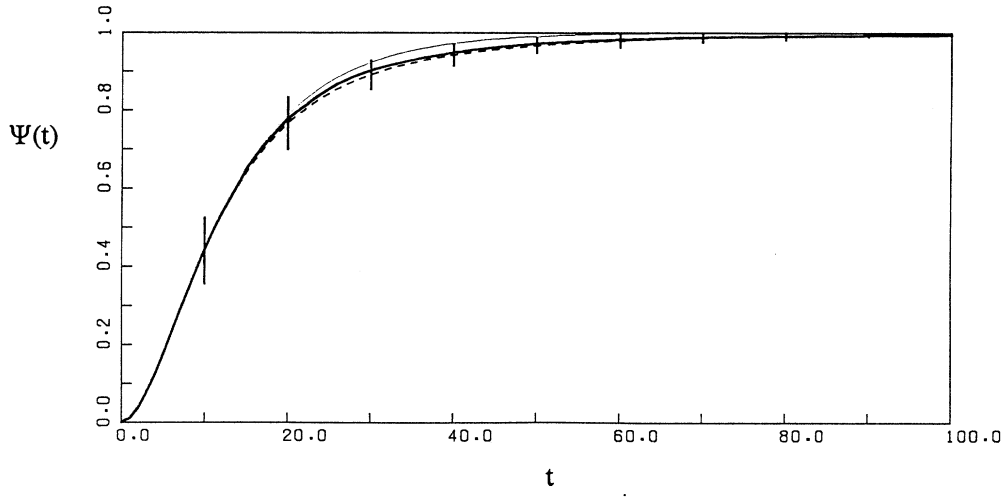

FIG. 7. The time dependence of the volume fraction for the decreasing growth rate. The meanings of the axes are the same as those in Fig. 4.

ing front velocity. The meanings of the lines are the same as those in Fig. 5. It is remarkable that the present theoretical line is almost overlapped with the average of the simulations.

In this way, it has been shown that the present theory, although not rigorous, provides us with a very accurate approximant for the volume fraction.

IV. CORRELATION FUNCTION OF THE ORDERED DOMAINS

As far as the total volume of the ordered domains is concerned, the degeneracy parameter p does not play any important role in the expression of the volume fraction. However, if we are interested in the structure of domains of a specific phase, i.e., the structure function, we need to determine the p dependence explicitly. In this section we aim to solve this problem in aribtrary dimensions under a condition as weak as possible.

We denote the ordered phases by $n=1,\ldots,p$ which are statistically equivalent. The metastable state is denoted by 0. The basic correlation function is $G_1(\mathbf{r}_1-\mathbf{r}_2,t)$ which is the probability such that both of the spatial points $\mathbf{r}_1$ and $\mathbf{r}_2$ are covered by the ordered domain 1 by time t after quench. Here we note that there are two possibilities to cover the points $\mathbf{r}_1$ and $\mathbf{r}_2$ by domain 1, as shown in Fig. 8. One is the case where these two points are covered by a common domain 1 [Fig. 8(a)] and the other is the case that the points are covered by domains 1 that have nucleated by different nucleation events [Fig. 8(b)]. Let us denote the former probability by $C_1(\mathbf{r}_1-\mathbf{r}_2,t)$. Notice that because of the equivalence of the degenerate ordered states the latter probability is the same as $H_1(\mathbf{r}_1-\mathbf{r}_2,t)$, the probability such that the point $\mathbf{r}_1$ is covered by domain one and the point $\mathbf{r}_2$ by domain 2 by time t. We further introduce the probability $C_0(\mathbf{r}_1-\mathbf{r}_2,t)$ that the points $\mathbf{r}_1$ and $\mathbf{r}_2$ do not belong to the metastable domain 0 at time t. The correlation function $G_1(\mathbf{r}_1-\mathbf{r}_2,t)$ then satisfies the relation

$$G_1(\mathbf{r}_1,\mathbf{r}_2,t)=\frac{p-1}{p}C_1(\mathbf{r}_1,\mathbf{r}_2,t)+\frac{1}{p^2}C_0(\mathbf{r}_1-\mathbf{r}_2,t)\ . \quad (4.1)$$

This can be proved as follows. First of all, it is obvious from the definitions that we have

$$G_1=C_1+H_1\ . \quad (4.2)$$

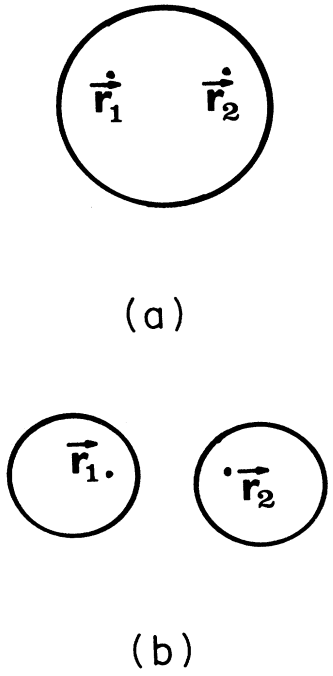

FIG. 8. Two possibilities to cover the points $\mathbf{r}_1$ and $\mathbf{r}_2$ by the same ordered phase. (a) The two points are covered by a common domain. (b) The two points are covered by two domains with different nucleation centers.

Here and after we omit the arguments $\mathbf{r}$ and t for simplicity. Next we note that the probability that the point $\mathbf{r}_1$ is covered by domain 1 and that the point $\mathbf{r}_2$ is covered by any of the ordered phases is given by $G_1+(p-1)H_1$. Hence we have

$$C_0=p\,[G_1+(p-1)H_1]\ . \tag{4.3}$$

Eliminating H_1 from (4.2) and (4.3), we obtain Eq. (4.1). It is noted here that another expression of C_0 is given by

$$C_0(\mathbf{r}_1-\mathbf{r}_2,t)=1-2\phi(t)+G_0(\mathbf{r}_1-\mathbf{r}_2,t)\ , \tag{4.4}$$

where G_0 is the probability that both points $\mathbf{r}_1$ and $\mathbf{r}_2$ belong to the metastable state at t.

The relation (4.1) was first obtained in Ref. 5 by a direct calculation of G_1 for spherical domains with a constant growth rate. It should be emphasized here that the present derivation indicates that (4.1) holds for more general conditions. What we have assumed is only the equivalence of the degenerate ordered phases.

We have attempted to evaluate explicitly the correlation functions G_1 and G_0 for the time-dependent growth rate by applying the method in Sec. II. However it turns out that the calculation is complicated. One difficulty is that we have to solve the coupled set of equations for G_1 and G_0 using the relation (4.1) generalized to two different times. Here we do not enter this problem further since we have not obtained any concrete solutions.

V. CONCLUDING REMARKS

We have investigated the domain growth with the time-dependent growth rate. The time evolution of the volume fraction has been obtained in one dimension. Here we discuss the possibility to generalize the theory to higher dimensions. When the growth rate is an increasing function of time, block of a growing domain by a newly formed domain occurs in one dimension as was shown in Sec. III. In higher dimensions, the situation is entirely different since a domain can grow around a new domain without interruption. Thus the block is less frequent unless the nucleation rate is too large. Only when a sufficient number of domains nucleate and surround a growing domain completely, the domain is blocked. This implies that the condition corresponding to no nucleation in the hatched region in Fig. 1 should be replaced by a more complicated one. Thus the theory for the increasing growth rate cannot be extended directly to higher dimensions. In the decreasing case, however, such a difficulty does not appear. We may apply the theory by using the higher-dimensional version of the causal cone in Fig. 3.

At the end of Sec. II, we have derived the volume fraction for the constant growth rate but with a finite nucleation radius. If we substitute the velocity (2.13) into the Kolmogorov-Avrami formula (2.4), we obtain

$$\psi(t)=1-\exp(-v_0I_0t^2-2I_0x_ct)\ . \tag{5.1}$$

This agrees with the result derived in Ref. 11 where (5.1) was obtained under the approximation, in the present terminology, such that the inside of the causal cone at time t_1 in Fig. 3 always belongs to the metastable state. The present result (2.14) is qualitatively different from (5.1). Especially when $v_0=0$, $\phi(t)$ shows an exponential decay in (5.1) while (2.14) exhibits a simple power-law decay $(x_cI_0t)^{-2}$.

So far we have considered the finite nucleation rate for $t>0$. It is mentioned that if the nucleation rate is given by $I(t)=I_0\delta(t)$, the factors $f(t_1,t_2)$ and $g(t_1,t_2)$ defined by (2.8) and (2.11), respectively, vanish trivially. In this case, the Kolmogorov-Avrami theory is valid for the time-dependent growth rate.

Computer simulations in Sec. IV show excellent agreement with the theory for both the increasing and the decreasing growth rates. Thus the theory, though here restricted to one dimension, will be useful for analyzing experimental observation such as in Ref. 10.

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