Dynamic scaling law for a first-order phase transition

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Giving a statistical-mechanical formulation of structure-function dynamics, we present a formulation of a scaling law for the first-order phase transition. The basic idea proposed, which previously appeared in a specific form, is that any function of the form $F_k(t_1,t_2,\ldots)$, where k is the wave number and $t_1,t_2,\ldots$ are times, is scaled as

$$F_k(t_1,t_2,\ldots)=[R(t_1)]^{x_1}[R(t_2)]^{x_2}\cdots\tilde{F}(kR(t_1),kR(t_2),\ldots)\ ,$$

where $R(t)$ is a relevant scale length, such as a linear dimension of an average cluster size, which is found to behave as $R(t)\propto t^{1/z}$ with z being a constant. The autocorrelation function of the density fluctuation is found to obey the scaling law for the conserved system:

$$J_k(t,t')=[R(t)]^{\theta}[R(t')]^{d-\theta}\tilde{J}(kR(t),kR(t')),\ \theta\leq d/2$$

where d is the dimensionality. For $\theta=0$ this scaling law can be naturally derived on the basis of the dynamic-scaling assumption and the conservation law. However, for large t the possibility of an anomalous scaling law ($\theta\neq 0$) is found. On the computer simulation for the three-dimensional spin-exchange kinetic Ising model we examine such a scaling law for the autocorrelation function. A remarkable difference in the temporal behaviors of the autocorrelation function is found. That observation strongly suggests the existence of the spinodal-like line.

I. INTRODUCTION

The classical theory of the spinodal decomposition founded by Cahn and Hilliard[1] and formulated by Cook[2] gave an impact on the later theoretical development[3–10] in the spinodal decomposition. The equation of motion for the time-dependent structure factor, which is often called the structure function, $S_k(t)$ takes the form

$$\frac{d}{dt}S_k(t)=ak^2[1-(bk^2-c)S_k(t)]$$

in the classical theory,[2] where a, b, and c are positive constants and k is a wave number. In the later works, the problem is how to determine a, b, and c, which may have dependences on k. The quantity a, b, or c in all later theories is time-dependent. This has been a main criticism to the classical theory. The theory for the intermediate stages of the spinodal decomposition by Langer, Bar-on, and Miller[4] gives good agreement with a computer simulation near the critical point.[11] The effect of the hydrodynamic force was treated as a perturbation to their theory by Kawasaki and Ohta,[7] which nicely explained the experiment of fluid mixture.[12]

In recent years, the scaling properties of the structure function of phase-separating binary mixtures in a late stage of spinodal decomposition have been studied theoretically,[6,8,13–15] by computer simulations[16,17] and experimentally.[18–22] In these works, attention is mainly focused on the scaling properties of the structure function

$$S_k(t)=[R(t)]^d\tilde{S}(kR(t))\ , \tag{1}$$

where $R(t)$ is a time-dependent characteristic length scale and d is the dimensionality. The exponent d is due to the saturation of composition in each phase in late stages of phase separation. The characteristic length scale R is proportional to the cluster size or the inverse of the peak position k_m of $S_k(t)$. The validity of the scaling law (1) is based on the fact that a relevant length scale is a linear dimension of cluster size R only, in the late stage of the decomposition where R is much larger than the thermal correlation length ξ. Theoretically, such a scaling idea appeared implicitly in the cluster-diffusion-reaction model by Binder and Stauffer.[6,13] The Lifshitz-Slyozov process of a binary alloy[23] with off-critical concentrations and Siggia's phenomenological model[24] for liquid mixtures with critical concentration are based on the similar kind of scaling ideas. The scaling form (1) was used by the present

author in order to analyze the equation of motion for $S_k(t)$.[8,14] The good agreements with low-temperature computer simulation[11] and an off-critical concentration fluid mixture[25] were obtained, and also experimental data[26] of binary alloy were analyzed. In all theoretical studies the scaling assumption (1) accompanies a simple power law for the temporal change of $R(t)$:

$$[R(t)]^z = At \ , \tag{2}$$

where A and z are constants. Recently, Rikvold and Gunton[15] calculated the scaling function $\tilde{S}(x)$ without the dynamical information and compared it with experimental data.[16,18]

It seems that we are now in a position to consider the scaling law for the first-order phase transition from more general viewpoint in order to discuss the scaling properties of other quantities. The purpose of this paper is to develop a scaling law for the dynamics of the first-order phase transition, in a somewhat general form, which already appeared in a specific form in previous work,[8,14] on the basis of nonequilibrium statistical mechanics. The scaling property of a two-time correlation function is one of many interesting subjects. Billotet and Binder[27] calculated the two-time correlation function by extending the theory of Langer, Bar-on, and Miller.[4] Their theory does not, however, satisfy the scaling law (1). The scaling property of a two-time correlation function or the autocorrelation function will be discussed generally. It seems that there is a difficulty arising from the uncertainty principle for examining a two-time correlation function using a differential cross section of particle or light scattered from a system in a time-dependent state. On the other hand, with a computer simulation we can develop a two-time correlation function without any difficulty. Therefore, instead of the theoretical example we shall study the two-time correlation function on a computer simulation. Two kinds of scaling law for the autocorrelation function in the system with conserved particle number are possible, i.e., one is a normal scaling law and the other is an anomalous scaling law. The latter may come out for large t or for large scale length R. An interesting classification of the mechanisms of the spinodal decomposition will be found from the behaviors of the autocorrelation function.

The basic statistical-mechanical method will be presented in Sec. II. We shall give a set of exact linear nonstationary equations for the density fluctuations, the autocorrelation function, and the structure function. The exact equation of motion for the structure function, which has the same kernel as that of the density fluctuation and the autocorrelation function, has not been derived so far. In Sec. III the dynamic scaling law for the first-order phase transition will be discussed. The scaling law for the autocorrelation function for the system with conserved particle numbers will be discussed. In Sec. IV the computer simulation for the three-dimensional spin-exchange kinetic Ising model is presented. Section V will be devoted to the discussions. The possibility of experimental observation of the scaling law for the autocorrelation function is also discussed. In Sec. VI summary and remarks will be presented.

II. STATISTICAL MECHANICAL PRELIMINARY

In previous papers[28] we have derived a formally rigorous Langevin-Mori–type equation[29] for a set of fluctuations of dynamical variables in nonequilibrium states. Such an equation is convenient to the present purpose, since in the spinodal decomposition the density fluctuations in the wave-number representation have zero average. The exact equation of motion for the density fluctuation n_k of minority phase (no essential difference exists if we discuss composition fluctuation instead of n_k as used previously[8,14]) is written as

$$\frac{d}{dt} n_{\vec{k}}(t) = K_k(t,t) n_{\vec{k}}(t) - \int_s^t \varphi_k(t,\tau) n_{\vec{k}}(\tau) d\tau + f_{\vec{k}}(t,s) \ , \tag{3}$$

$$\langle f_{\vec{k}}(t,s) n_{-\vec{k}}(s) \rangle_0 = 0, \quad t \geq s \tag{4}$$

where $\langle \ \rangle_0$ means an ensemble average in a nonequilibrium state, and

$$K_k(t,t) \equiv \langle \dot{n}_{\vec{k}}(t) n_{-\vec{k}}(t) \rangle_0 S_k(t)^{-1} \ . \tag{5}$$

Here the dot stands for time derivative and $S_k(t)$ is the structure function

$$S_k(t) = \langle n_{\vec{k}}(t) n_{-\vec{k}}(t) \rangle_0 \ . \tag{6}$$

Notice that $\langle n_{\vec{k}}(t) \rangle_0 = 0$ for $k \neq 0$ even in the nonequilibrium state. The memory function $\varphi_k(t,\tau)$ is related to the fluctuating force with a generalized fluctuation-dissipation theorem[28,30]

$$\varphi_k(t,\tau) S_{\vec{k}}(\tau) - \langle f_{\vec{k}}(t,\tau) f_{-\vec{k}}(\tau,\tau) \rangle_0 = 0 \ . \tag{7}$$

The equation of motion for the autocorrelation function defined by

$$J_k(t,s) \equiv \langle n_{\vec{k}}(t) n_{-\vec{k}}(s) \rangle_0 \ , \tag{8}$$

which is a real quantity as well as $S_k(t)$ due to the spatial symmetry, is obtained from (3) and (4) as

$$\frac{d}{dt}J_k(t,s)=K_k(t,t)J_k(t,s)-\int_s^t\varphi(t,\tau)J_k(\tau,s)d\tau$$
$$=\int_s^t\Theta_k(t,\tau)J_k(\tau,s)d\tau\ , \tag{9}$$

where

$$\Theta_k(t,\tau)\equiv 2K_k(t,t)\delta(t-\tau)-\varphi_k(t,\tau)\ . \tag{10}$$

We shall here derive an exact equation of motion for $S_k(t)$, which has not been derived so far. Using (5) we have

$$\frac{d}{dt}S_k(t)=[K_k(t,t)+K_k^*(t,t)]S_k(t)$$
$$=2K_k(t,t)S_k(t)\ , \tag{11}$$

where the asterisk denotes the complex conjugate. $K_k(t,t)$ is also real by the reason mentioned above. By integrating (7) over τ in the region from s to t, and subtracting the resultant equality and its complex conjugate from the second side of (11), we obtain

$$\frac{d}{dt}S_k(t)=\int_s^t[\Theta_k(t,\tau)+\Theta_k^*(t,\tau)]S_k(\tau)d\tau$$
$$+N_k(t,s)+N_k^*(t,s)\ , \tag{12}$$

where

$$N_k(t,s)=\int_s^t\langle f_{\vec{k}}(t,\tau)f_{-\vec{k}}(\tau,\tau)\rangle_0 d\tau\ . \tag{13}$$

Equation (12) as well as (9) is an identity and therefore is a rigorous equation of motion for $S_k(t)$. The essential point is that (12) has the same memory kernel as that of (9) or (3). Therefore, this provides us with a rigorous extension of the variance equation obtained by the system-size expansion.[31,32]

For the scaling analysis of the spinodal decomposition, a relatively large time scale such as a time in which average cluster changes its size by the amount of the order of itself is important. The assumption that the fluctuating force has a short-time memory may not lose its generality of the following discussions. Mathematically this can be done by setting

$$\Theta_k(t,\tau)=-2\Gamma_k(t)\delta(t-\tau)\ . \tag{14}$$

Then (3), (9), and (12) reduce, respectively, to

$$\frac{d}{dt}n_{\vec{k}}(t)=-\Gamma_k(t)n_{\vec{k}}(t)+f_{\vec{k}}(t)\ , \tag{3'}$$

$$\frac{d}{dt}J_k(t,s)=-\Gamma_K(t)J_k(t,s)\ , \tag{9'}$$

and

$$\frac{d}{dt}S_k(t)=-2\Gamma_k(t)S_k(t)+2N_k(t)\ , \tag{12'}$$

where

$$N_k(t)=\int_{t-0}^t\langle f_{\vec{k}}(t)f_{-\vec{k}}(t')\rangle_0 dt'$$
$$\simeq\int_{t-0}^t\langle\dot{n}_{\vec{k}}(t)\dot{n}_{-\vec{k}}(t')\rangle_0 dt'\ . \tag{13'}$$

Equation (12') has the same form as that of variance equation for the system-size expansion[31,32] and also has been used for the study of the spinodal decomposition.[8,14]

When (9') and (12') are applied to the second-order phase transition,[33] Γ_k, S_k, and N_k are time independent. Then (12') gives the usual fluctuation-dissipation relation. The static and dynamic scaling assumption for the second-order phase transition are written as

$$S_k\equiv\chi_k=\xi^{z_0-z_0'}\tilde{\chi}_0(k\xi)\ , \tag{15}$$

$$\Gamma_k=k^{\xi_0}\tilde{\Gamma}_0'(k\xi)\ , \tag{16}$$

where z_0 and z_0' are constants. We also observe that

$$N_k=\Gamma_k\chi_k=k^{z_0'}\tilde{N}'(k\xi)\ . \tag{17}$$

III. DYNAMIC SCALING LAW FOR SPINODAL DECOMPOSITION

For the first-order phase transition S_k, Γ_k, and N_k are time dependent. We shall now formulate the scaling law for the spinodal decomposition. The scaling assumption for the spinodal decomposition is that all such quantities are scaled by a single time-dependent scale length R and depend time only through R:

$$\Gamma_k(t)=[R(t)]^{-z}\tilde{\Gamma}(kR(t))$$
$$=k^z\tilde{\Gamma}'(kR(t))\ , \tag{18}$$

$$N_k(t)=[R(t)]^{-z'}\tilde{N}(kR(t))$$
$$=k^{z'}\tilde{N}'(kR(t))\ , \tag{19}$$

where $\tilde{\Gamma}'(x)=x^{-z}\tilde{\Gamma}(x)$ and $\tilde{N}'(x)=x^{-z'}\tilde{N}(x)$. In order that (12') has a scaling solution (1), it follows that

$$\frac{d}{dt}R^d\propto R^{-z+d}\propto R^{-z'}\ . \tag{20}$$

Thus, (2) follows together with

$$z-z'=d\ . \tag{21}$$

Here z in (18) is initially independent of z in (2). But now it becomes apparent that both exponents are equivalent.

The scaling law (18) and (19) may be generalized for the case where the memory kernel $\varphi(t,\tau)$ has a finite memory. They are written as

$$K_k(t,t)=[R(t)]^{-z}\tilde{K}(kR(t)) , \tag{18'}$$

$$\varphi_k(t,\tau)=[R(t)]^{-z}[R(\tau)]^{-z}\tilde{\varphi}(kR(t),kR(\tau)) , \tag{18''}$$

$$\langle f_k(t,\tau)f_{-k}(\tau,\tau)\rangle_0=[R(t)]^{-z'}[R(\tau)]^{-z}\tilde{M}(kR(t),kR(\tau))=[R(t)]^{-z}[R(\tau)]^{-z'}\tilde{M}'(kR(t),kR(\tau)) . \tag{19'}$$

Here from (7) we have

$$\tilde{\varphi}(x,y)\tilde{S}(y)=\tilde{M}'(x,y)\equiv(x/y)^d\tilde{M}(x,y) . \tag{22}$$

Equations (2) and (21) also follow. The scaling function $\tilde{\Gamma}$ and $\tilde{N}$ in (18) and (19) are related to $\tilde{K}$, $\tilde{\varphi}$, and $\tilde{M}$ as follows:

$$\tilde{\Gamma}(x)=-\tilde{K}(x)+A^{-1}z\int_0^x\tilde{\varphi}(x,y)\frac{dy}{y} , \tag{23}$$

$$\tilde{N}(x)=A^{-1}z\int_0^x\tilde{M}(x,y)\frac{dy}{y} . \tag{24}$$

The equation of motion (12') is rewritten as

$$\frac{d}{dt}S_k(t)=2N_k(t)[1-\chi_k^{-1}(t)S_k(t)] , \tag{25}$$

where

$$\chi_k(t)\equiv N_k(t)/\Gamma_k(t)=R^d\tilde{\chi}(kR)=k^{-d}\tilde{\chi}'(kR) . \tag{26}$$

Thus, χ_k has the same scaling exponent d as $S_k(t)$. χ_k corresponds to the susceptibility in the equilibrium statistical mechanics. By solving (9') and (12') we obtain

$$J_k(t,t')\equiv\langle n_k(t)n_{-k}(t')\rangle_0=U_k(t,t')S_k(t') , \tag{27}$$

$$S_k(t)=[U_k(t,t_0)]^2S_k(t_0)+[U_k(t,t_0)]^2\times\int_{t_0}^t[U_k(\tau,t_0)]^{-2}N_k(\tau)d\tau , \tag{28}$$

where

$$U_k(t,t_0)\equiv\exp\left[-\int_{t_0}^t\Gamma_k(\tau')d\tau'\right]=\exp\left[-\frac{z}{A}\int_{R(t_0)}^{R(t)}\tilde{\Gamma}(kR')\frac{dR'}{R'}\right]=\exp\left[-z\int_{R(t_0)}^{R(t)}\tilde{\Gamma}^*(kR)\frac{dR'}{R'}\right] \tag{29}$$

and where

$$\tilde{\Gamma}^*\equiv A^{-1}\tilde{\Gamma} .$$

We can find that

$$\Gamma_k(t)=t^{-1}\tilde{\Gamma}^*(kR(t)) .$$

We have used (2) to transform the second side of (29) into the third side. t_0 is some constant, which is introduced in order to avoid any difficulty in dealing with trivial small scale length $R<R(t_0)$.

We shall here derive the scaling law for the autocorrelation function, for the system with conserved particle number. We shall show that there are two possibilities for the scaling law for the autocorrelation function. The one is more general than the other. However, the general one is anomalous from the viewpoint of the dynamic scaling law as will be seen below. Such an anomalous scaling law is not, however, ruled out.

A. Normal scaling

For a system with conserved particle numbers, Γ_k should be zero at $k=0$ since (3') is a diffusion equation:

$$\lim_{k\to 0}\Gamma_k=0 . \tag{30}$$

Therefore, it is natural to assume that the exponential factor of (29) does not have a logarithmic term, and we therefore obtain a *normal* form of the scaling law for $U_k(t,t')$:

$$U_k(t,t')=V_k(t)[V_k(t')]^{-1} , \tag{31}$$

$$V_k(t)=\tilde{V}(kR(t)) . \tag{32}$$

Thus, the scaling law for the autocorrelation function is written from (27) as

$$J_k(t,t')=[R(t')]^d\tilde{J}(kR(t),kR(t')) , \tag{33}$$

where

$$\tilde{J}(x,x')=\tilde{V}(x)[\tilde{V}(x')]^{-1}\tilde{S}(x') . \tag{34}$$

For $R(t)\gg R(t')$ we have

$$J_k(t,t')\simeq J_k(t,0)\simeq\tilde{J}(kR(t)) . \tag{33'}$$

B. Anomalous scaling

The scaling law (33) or (33') is indirect to the scaling law (1) which seems to have a firm physical basis as discussed in Sec. I. Although the conservation law plays a fundamental role when we derive (33) or (33'), this is not enough to avoid the possibility of the other scaling law different from (33) or (33'). In fact, a rigorous restriction to the autocorre-

lation function is only the Schwartz inequality:

$$|J_k(t,t')|^2 \leq S_k(t)S_k(t') .$$

Therefore, it must be worthwhile to examine the possibility of another scaling law for the autocorrelation function. We shall show that the following scaling law for $V_k(t)$ as

$$V_k(t)=[R(t)]^{\theta}\tilde{V}(kR(t)), \quad \theta\neq 0 \tag{32'}$$

is also possible. From (1), (27), (31), and (32') we have an anomalous scaling law for $J_k(t,t')$:

$$J_k(t,t')=[R(t)]^{\theta}[R(t')]^{d-\theta}\tilde{J}(kR(t),kR(t')) , \quad \theta\neq 0 . \tag{35}$$

For $R(t)\gg R(t')$, (36) may be approximated as

$$J_k(t,t')\simeq J_k(t,0)\simeq[R(t)]^{\theta}\tilde{J}(kR(t)) . \tag{35'}$$

From (35') and the above Schwartz inequality we find that

$$\theta\leq d/2 . \tag{36}$$

From (29) and (32'), $\Gamma_k(t)$ in this case is found to be

$$\Gamma_k(t)=-\theta z^{-1}t^{-1}-\frac{d}{dt}\ln\tilde{V}(kR(t)) . \tag{37}$$

Thus, the damping coefficient even for the conserved system has a constant term in this case. This is the reason why we call the scaling law (32') for the conserved system an anomalous scaling law.

We shall here show a trick to get the anomalous scaling law (32') for the conserved system. From the conservation law, $V_k(t)$ must have an expression for small k,

$$V_k(t)=\exp[\alpha(t)k^m], \quad m>0 \tag{38}$$

which is not yet scaled. Equation (38) is now rewritten as

$$V_k(t)=R^{\theta}\exp[-\ln R^{\theta}+\alpha(t)k^m] . \tag{38'}$$

Then we may think that the exponential factor of (38') is the lowest order of the expansion of a complete expression such as

$$V_k(t)=R^{\theta}\exp[-1/(\alpha' x^m+1/\ln R^{\theta})] ,$$
$$x\equiv kR(t), \quad \alpha(t)=\alpha' R^m(\ln R^{\theta})^2 , \tag{38''}$$

for not too large k or x, where α' is a constant independent of time. For the present example (38''), the scaling law (32') holds for $\alpha' x^m>1/\ln R^{\theta}$, while the scaling law (32') or (32) will not hold for $\alpha' x^m<1/\ln R^{\theta}$. The scaling expression for $V_k(t)$ is therefore

$$V_k(t)=R^{\theta}\exp(-1/\alpha' x^m), \quad \alpha' x^m>1/\ln R^{\theta} .$$

The corresponding expression for $\Gamma_k(t)$ is given from (37) as

$$\Gamma_k(t)=-(\theta+m/\alpha' x^m)/zAR^z , \quad \alpha' x^m>1/\ln R^{\theta} .$$

The scaling law (32') thus holds for large t, i.e., for large R. The above discussion is by no means a rigorous one, but gives an example of the anomalous scaling law for the autocorrelation function for the system with conserved particle numbers. There is no reason why we deny the existence of the logarithmic term in the integration of the second term of (28). If the logarithmic term exists, then the second term of (28) gives the contribution of the form $(R^d\ln R)\tilde{S}_1(kR)$. However, such a contribution gives no serious deviation from the scaling law (1). On the other hand, a similar kind of logarithmic term in the exponential factor of (29) gives a drastic change in the scaling law for $J_k(t,t')$. From this fact we know that the scaling law (1) is more stable than the scaling law (33) or (35). We also consider that the scaling law (1) would be approximately correct even if the scaling law (33) or (35) does not hold.

Although the scaling theory discussed here is basically the direct extension of the field-theoretical approach in previous work,[8,14] this is also closely related to the phenomenological scaling theories.[13,23,24] We should, however, be careful to deal with phenomenological models. Since, the phenomenological modes do not explicitly treat the contribution from the fluctuating force, i.e., $N_k(t)$. $N_k(t)$ always has the contribution from the bare thermal noise,[2] i.e.,

$$N_k^0=k_B TM^0k^2 \tag{39}$$

which can be scaled as

$$N_k^0=R^{-2}k_B TM^0(kR)^2 . \tag{40}$$

Here M^0 is a constant. If N_k^0 is only the contribution from the fluctuating force, we then obtain[8]

$$a'=a_0'\equiv(d+2)^{-1}, \quad z=z_0\equiv d+2 ,$$
$$z'=z_0'\equiv 2 ,$$

where $a'\equiv d\ln R/dt=1/z$. In the phenomenological models, not z' but a' is computed. The contribution from the bare fluctuating force is not treated in phenomenological models. Thus, by letting a_1' be the exponent obtained by a phenomenological model, one may have[14]

$$N_k(t)=N_k^0+N_k^1(t) \tag{41}$$

with

$$N_k^1(t)=R^{-z_1'}\tilde{N}^1(kR), \quad z_1'=1/a_1'-d . \tag{42}$$

By comparing (42) with (40) we can find that N_k^1 can be neglected for $z_1' > z_0'$ or $a_1' < a_0' = 1/(d+2)$ (for large t), while N_k^0 can be neglected for $z_1' < z_0'$ or $a_1' > a_0'$. This means that $1/(d+2)$ gives a lower limit of the exponent a' in the field-theoretical method. The cluster-diffusion-reaction model by Binder and Stauffer[13] gives $a_1' = 1/(d+3)$ for the spin-exchange kinetic Ising model at low temperatures. In the field-theoretical method, such a reaction process should be neglected and the exponent a' should not be $1/(d+3)$, but $1/(d+2)$.[14] On the other hand, the cluster-diffusion-reaction model for fluid mixture with off-critical concentration gives $a_1' = 1/d$.[13] In this case N_k^0 should be neglected for large R, and the field-theoretical method[14] gives the same exponent $a' = a_1' = 1/d$ as that of the cluster-diffusion-reaction model. As can be found from the above discussion, for $a_1' \geq 1/(d+2)$ we always expect that the phenomenological exponent a_1' is equivalent to the field-theoretical one. Therefore, for both the Lifshitz-Slyozov process,[23] where $a_1' = \frac{1}{3}$, and for Siggia's process,[24] where $a_1' = 1$, the field-theoretical approach should give the same exponents as phenomenological ones, i.e., $a' = a_1'$. The exponent $a' = 1/(d+2)$ at low temperatures is due to the individual transfers of atoms, while the phenomenological exponents are due to the cooperative transfers of atoms.

There is a similarity between the scaling law for the second-order phase transition[33] and that for the spinodal decomposition. The damping coefficient Γ and the strength of the fluctuating force N are scaled in similar ways in both cases by a single scale length, i.e., the thermal correlation length ξ and the linear dimension of the average size of droplet R, respectively. Such a similarity was also noted by Binder,[6] regarding the diffusion constants of droplets in both cases. Some important differences should, however, be recognized. Almost all exponents in both cases are not related to each other. Especially, the exponent d for $S_k(t)$ and $\chi_k(t)$ in the spinodal decomposition does not hold for the second-order phase transition. All corresponding functions are different from each other. For instance, $\tilde{\chi}(x)$ or $\tilde{S}(x)$ does not have x^{-2} tail in the spinodal decomposition. It is usually considered that $\tilde{S}(x)$ has $x^{-\gamma}$ tail with $d+1 \leq \gamma \leq 2d$.[8,14] γ depends on the surface condition of droplets. If the surfaces of droplets are not so tangled, then $\gamma = d+1$. Such an x^{-d-1} tail was ascertained by Lebowitz *et al.*[16] The scattering cross section of fluid mixture with off-critical concentration[25] also seems to show such a tail as k^{-4}. x^{-d-1} tail was discussed also by Rikvold and Gunton.[34] Such an x^{-d-1} tail of $\tilde{S}(x)$ is also found for the system with nonconserved order parameter,[34] which has the same origin as that of the conserved system.

The most important difference between the scaling laws of the second-order phase transition and the first-order phase transition is the following. In the first-order phase transition no final state is reached in a finite time, while all phenomena occur around the final state in the second-order phase transition. This makes the two problems almost uncorrelated. In the second-order phase transition the autocorrelation function $J_k(t,0)$ explicitly contains time t. However, the inverse of time t^{-1} in the spinodal decomposition plays as the temperature $T - T_c$ in the second-order phase transition.

V. COMPUTER SIMULATION

We shall present here results of computer simulation on a three-dimensional spin-exchange kinetic Ising model.[35] The main purpose of our simulation is not to examine the scaling law (1), but to examine the scaling law for $J_k(t,0)$ [see Eq. (35′)]. The scaling law for $S_k(t)$ was extensively studied by Lebowitz, Marro, and Kalos.[16] The purpose of studying $S_k(t)$ is to understand the time dependence of $R(t)$ in the present simulation. The simulation was done in a similar way as in previous work.[36] A system is suddenly quenched from a state with initial random configuration into a two-phase region. The system has $24 \times 24 \times 24$ lattice sites with simple cubic lattice structure. Monte Carlo samplings are, however, done not for each lattice site, but for each particle of a minority species, in order to gain a computation time (we made simulation for small densities less than 0.1). Usually Monte Carlo samplings are made for lattice sites. The meaningful exchanges are, however, those between different species. This fact makes two random sampling methods essentially equivalent.

$S_{\vec{k}}(t)$ and $J_{\vec{k}}(t,0)$ are computed to

$$S_{\vec{k}}(t) = \frac{1}{N} \sum_{\vec{r}} \sum_{\vec{r}_i} [n(\vec{r}_i + \vec{r}, t) n(\vec{r}_i, t) - \bar{n}^2] \times e^{i \vec{k} \cdot \vec{r}}, \tag{43}$$

$$J_{\vec{k}}(t,0) = \frac{1}{N} \sum_{\vec{r}} \sum_{\vec{r}_i} [n(\vec{r}_i + \vec{r}, t) n(\vec{r}_i, 0) - \bar{n}^2] e^{i \vec{k} \cdot \vec{r}}, \tag{44}$$

where $\vec{r}$ and $\vec{r}_i$ run over $N = 24^3$ sites and $n(\vec{r},t)$ is $+1$ if site $\vec{r}$ is occupied, and 0 if otherwise. $\bar{n}$ is the average density, i.e., $\bar{n} = N_{\text{tot}}/N$, where N_{tot} is the total particle number of minority phase, which is conserved. $\vec{K}$ is a discrete wave-number vector

$$\vec{k} = (\tfrac{2}{24}\pi)(\mu_x, \mu_y, y_z) \tag{45}$$

with $\mu_\alpha=0,1,..,23$ and $\alpha=x,y,z$. For definition, $S_{\vec{k}}(t)\simeq\bar{n}$ for $\vec{k}\neq 0$. The most interesting quantities are spherically averaged ones:

$$S_k(t)=\sum_{\vec{k}}{}' S_{\vec{k}}(t)\Big/\sum_{\vec{k}}{}' 1 , \tag{46}$$

$$J_k(t,0)=\sum_{\vec{k}}{}' J_{\vec{k}}(t,0)\Big/\sum_{\vec{k}} 1 , \tag{47}$$

where $k=(\frac{2}{24})\pi\mu$, $\mu=1,2,\ldots,$ and the sum $\sum'$ goes over all values of μ such that

$$(\tfrac{2}{24})\pi\mu\leq|\vec{k}|<(2/24)\pi(\mu+1) .$$

All these definitions except for $J_k(t,0)$ are the same as that of Ref. 11. The final form for $S_k(t)$ and $J_k(t,0)$ are obtained by averaging over eight different runs with the same density and the same temperature T. We have done the simulation at $T=0.44T_c$ [case (a)] and $T=0.59T_c$ [case (b)] with the average density $\bar{n}=0.075$ (Figs. 1 and 2). The time interval for simulations is $0\sim 8000$ in both cases, where one try per particle of minority species is taken as unit time. Our results for $S_k(t)$ are consistent with those of Ref. 16. We have obtained the exponents z and θ in each case as

$$1/z\simeq 0.2,\quad \theta\simeq 0$$

$$1/z\simeq 0.33,\quad \theta\lesssim 0$$

in cases (a) and (b), respectively. Here the exponent z's were given so as to get a smooth function $\tilde{S}(x)$, and θ's were given to get $\tilde{J}(x)$. However, these evaluations may not be the most suitable ones. Slight changes in these values do not yield remarkable changes in the scaling function $\tilde{S}(x)$ and $\tilde{J}(x)$.

I. DISCUSSION

It might be found that $J_k(t,0)$ becomes slightly negative at certain wave numbers. Since $J_k(t,0)$ is real, $\Gamma_k(t)$ in (9′) should be real. This means that $J_k(t,0)$ in (9′) cannot be negative. This discrepancy between the theoretical prediction and experimental observations is due to the short-time approximation for the memory of the fluctuating force in the theory. The memory effect should be considered. Since, however, we have not observed such a memory effect for $S_k(t)$, the memory effect would be small.

In both cases of the simulations we may observe that the peak position of $J_k(t,0)$ saturates after ending a relatively short transient state, thus satisfying the normal scaling law (33). However, there is a remarkable difference between the behavior of the autocorrelation functions in both cases. Namely, the peak of the autocorrelation functions in case (a) rapidly grows after the quench, while that in case (b) rapidly decreases after the quench. In both cases the structure function grows, satisfying the scaling law (1), as already shown extensively by Lebowitz *et al.*[16] In case (a) we therefore find that a part of the damping coefficient $\Gamma_k(t)$ is negative as was traditionally believed. On the other hand, in case (b) there is no negative part in the damping coefficient $\Gamma_k(t)$. This seemingly peculiar phenomenon, which was, however, previously predicted for

$$\chi_k(t)\equiv N_k(t)\Gamma_k(t)^{-1}$$

(Ref. 14), may come from the fact that each droplet behaves as a free-Brownian movement, which wanders faster than its growth rate. Then the Brownian movements easily lose their memories of positions. Thus, $J_k(t,0)$ may decrease, i.e., Γ may be positive. In this case, $N_k(t)$ is responsible to the growth of the structure function

$$S_k(t)\simeq\begin{cases}\int_0^t N_k(t')dt', & \text{for } k\lesssim k_m \quad (48a)\\ \chi_k(t), & \text{for } k>k_m . \quad (48b)\end{cases}$$

That is, $S_k(t)$ is nearly independent of Γ_k for $k\lesssim k_m$. In such a case we are anxious about the validity of the scaling law (33) or (35) for the autocorrelation function, since the scaling law (1) is independent of scaling law (33) or (35). Furthermore, since $J_k(t,0)$ decays, the contribution from the microscopic degrees of freedom cannot be neglected. This also makes it difficult to justify the scaling law for the autocorrelation function in this case.

We should here remark on another of our observations. For a certain set of different runs for $T=0.44T_c$ and $\bar{n}=0.05$ we have observed the growth in the peak of the autocorrelation function, satisfying the anomalous scaling law (35′). But for another set of the runs for the same temperature and the same density we observed that the peak height of the autocorrelation function initially grew and then approximately saturated. In both cases the structure function $S_k(t)$ behaves almost in the same way. At this moment, therefore, we cannot conclude from our computer simulation which scaling law the autocorrelation obeys best. We consider that the anomalous scaling law for the autocorrelation function for the conserved system should not be ruled out. For instance, if the strength of the fluctuating force $N_k(t)$ is neglected for $k<k_m$, we then should have

$$S_k(t)\propto[J_k(t,0)]^2 \quad \text{for } k\lesssim k_m \tag{49}$$

and therefore $\theta=d/2$ follows, showing the anomalous scaling law for $J_k(t,0)$. Another consideration also suggests the possibility of the existence of the anomalous scaling law. A simple scaling analysis for the density fluctuation

$$n_k(t)=[R(t)]^{d/2}\tilde{n}(kR(t)) \tag{50}$$

leads to $\theta=d/2$. Equation (50) does not always hold, however, since an appropriate expression for $n_k(t)$ is

$$n_k(t)=U_k(t,t')n_k(t')+U_k(t,t')\int_{t'}^{t}[U_k(\tau,t')]^{-1}f_k(\tau)d\tau\ . \tag{51}$$

The second term gives the scaling law (1), even if θ is not equivalent to $d/2$. Much more information about the autocorrelation functions of phase-separating binary mixtures will be needed. Notice that the scaling law (35) or (35') is not anomalous if the order parameter is not conserved. We will consider here the physical meaning of the different behaviors in the autocorrelation functions on the above simulation. We can observe that the autocorrelation function $J_k(t,0)$ in case (a) initially increases, while that in case (b) initially decreases. This means that $\Gamma_k(t)$ has a negative part in case (a), while it has no negative part in case (b) in the early stage of the phase separation just after the quench. Since the damping coefficient $\Gamma_k(t)$ is directly related to the second derivative of the free energy, the second derivative of the free energy in case (a) has a negative part, while it has no negative part in case (b) in the early stage of the phase separation. This indicates that there is a spinodal-like line in between the two states considered above (see Fig. 3). This line should, however, be distinguished from the spinodal line in a usual sense. The spinodal-like line is a line where a free energy of metastable states becomes singular. That free energy is obtained from the analytic continuation or coarse graining. The spinodal-like line thus obtained is not unique, but depends on parameters such as a volume of the coarse-grained cell in a system with short-range interacting forces. It is plausible that the spinodal-like line identified in the present work is related to the spinodal-like line through an unknown volume size of the coarse-grained cell. At this moment, however, we can say nothing clear about the relation of the spinodal-like line identified in the present work with the spinodal line in a usual sense.

So far, to set a spinodal-like line by means of only the structure function has never been succeeded. This is because that the equation of motion for the structure function has an inhomogeneous term $N_k(t)$

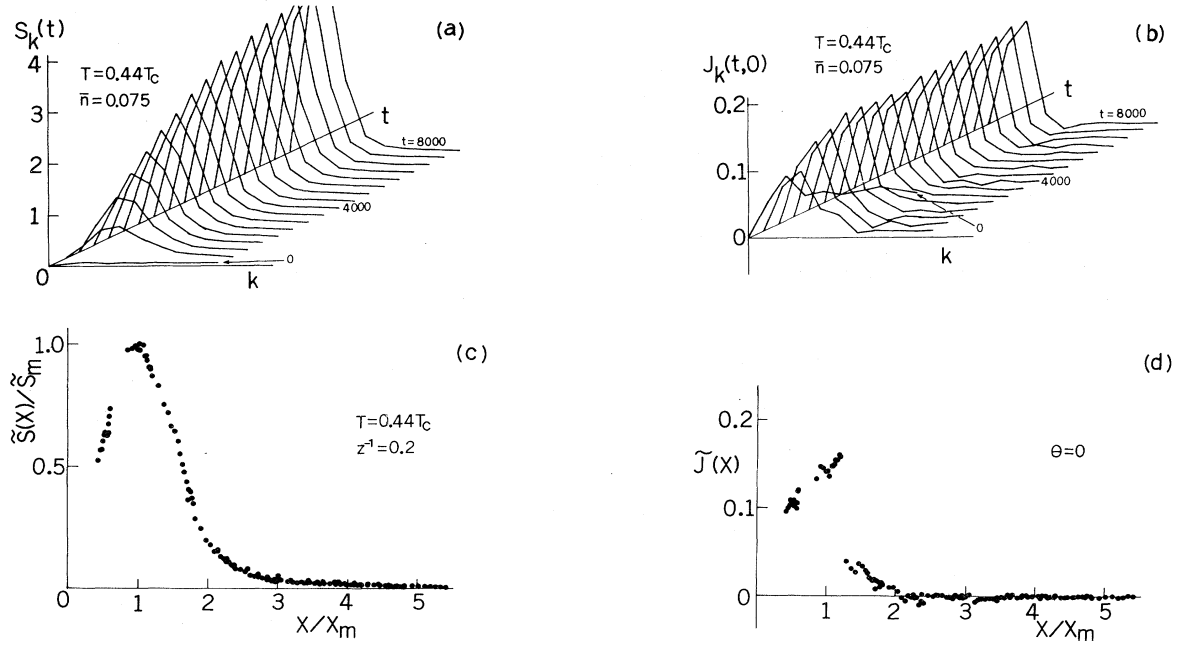

FIG. 1. (a) Structure function, (b) autocorrelation function, (c) and (d) scaling functions corresponding to (a) and (b), respectively, for $T=0.44T_c$, $\bar{n}=0.075$. Data for $1500\leq t\leq 8000$ are used for the scaling functions. Since $\theta=0$ and $J_k(0,0)=S_k(0)=0.075$, one may find that $\Gamma<0$ for $\tilde{J}>\bar{n}$.

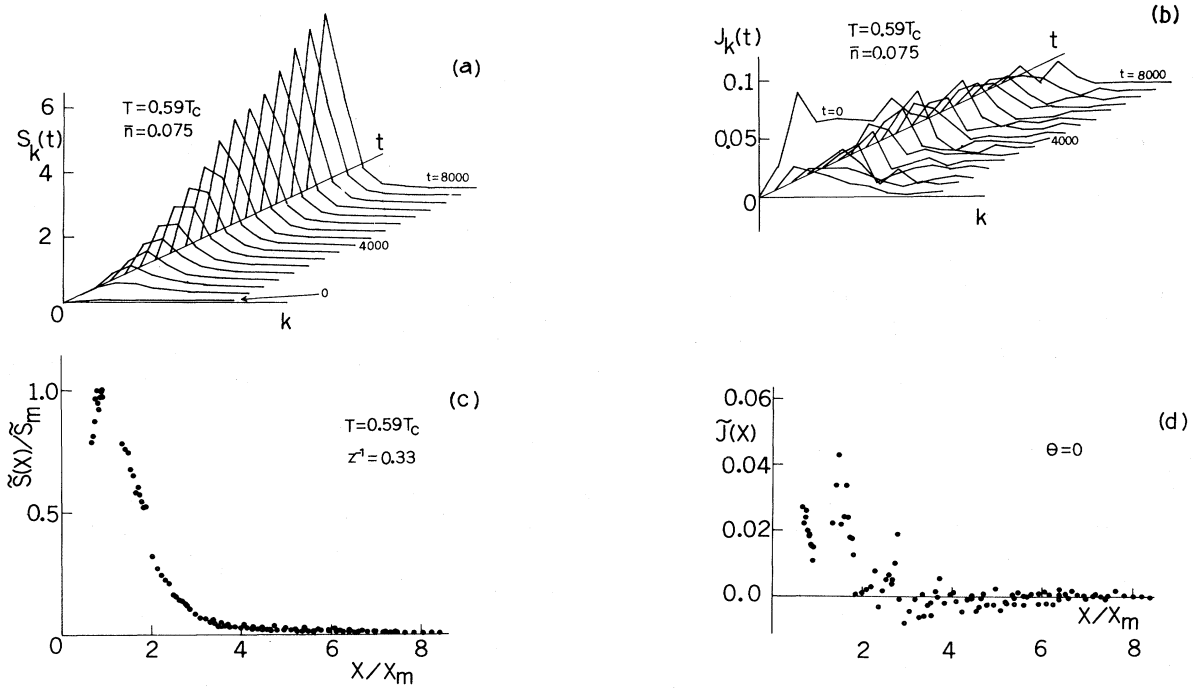

FIG. 2. (a)—(d) are the same as in Fig. 1 for $T=0.59T_c$ and $\bar{n}=0.075$. Data for $3000 \leq t \leq 8000$ are used for the scaling functions. Since $\tilde{J} < \bar{n}=0.075$ for all x, the damping coefficient Γ has no negative part.

[see (12′)], which masks the contribution from the damping coefficient $\Gamma_k(t)$. Therefore, the autocorrelation function is a powerful tool for the study of the mechanisms of the phase separations, e.g., to set a spinodal-like line, which may be determined from the initial behaviors of the autocorrelation function.

We finally consider the possibility of the experimental observation of the scaling law for the autocorrelation function. The differential cross section of particles scattered from a system in a nonequilibrium state is given as (the second reference of Ref. 28)

$$\sigma_{k,\omega}(t) \propto \mathrm{Re} \int_{-\infty}^{t} dt' e^{i\omega(t-t')} J_{\vec{k}}(t,t') \ . \tag{52}$$

Here $\hbar\omega$ are the energy transfers of scattered particles. Billotet and Binder[27] found that the scattering cross section (52) in the nonequilibrium state has negative values. They used $J_k(t,t')$ obtained from the theory of Langer, Bar-on, and Miller.[4] They considered that the negativeness of the differential cross section is due to the approximation used to obtain $J_k(t,t')$. Partly, this might be correct, since $J_k(t,t')$ has a negative part as was found in the present computer simulation. $J_k(t,t')$ calculated by Billotet and Binder does not become negative. However, this is not all for the negativeness of the calculated cross section. The negativeness of the theoretical cross section mainly comes from the violation of the uncertainty principle. In order to catch scattered particles with energy change $\hbar\omega$ the observation time ω^{-1} is needed at least. If we want to know the information of the system in the time interval τ, in which the state of the system remarkably changes, it is not meaningful to consider the scattering cross section of particles with energy change $h\tau^{-1}$. In an equilibrium state we can take τ as infinitely large, and the differential cross section does not become negative. Therefore, in order to apply the formula (52) to nonequilibrium states, $\sigma_{k,\omega}(t)$ should be replaced by the one averaged at least over the time interval ω^{-1}. We, however, consider that the scattering cross section (52) can be used for the study of the asymptotic behaviors of the differential cross section, i.e., the examination of the scaling law of the autocorrelation function. By substituting (35) into (52) we have

$$\sigma_{k,\omega}(t) \propto \mathrm{Re}[R(t)]^{\theta} \int_{-\infty}^{t} dt' e^{i\omega(t-t')} [R(t')]^{d-\theta} \tilde{J}(kR(t), kR(t')) \ . \tag{53}$$

If the integrand of (53) changes rapidly, then $\sigma_{k,\omega}(t)$ has a time dependence as

$$\sigma_{k,\omega}(t) \propto [R(t)]^{d} \ , \tag{54}$$

which is, however, essentially the same as that of the scattering cross section from an equilibrium system. On the other hand, if the integrand of (53) changes slowly, then $\sigma_{k,\omega}(t)$ has a time dependence, which is different from (54), as

$$\sigma_{k,\omega}(t) \propto [R(t)]^{\theta} \ . \tag{55}$$

Equation (55) provides us with a possibility to observe the scaling law for the autocorrelation function. The total cross section

$$\sigma_{k}(t) \propto \int_{-\infty}^{\infty} d\omega \, \sigma_{k,\omega}(t) \tag{56}$$

always behaves in the same way as (54).

VI. SUMMARY AND REMARKS

The main purpose of this paper has been to explore the general framework of the scaling law for the first-order phase transition. An exact description of the motion for the structure function $S_k(t)$ and the autocorrelation function $J_k(t,t')$ are available. The equations of motion for both quantities have the same memory kernel.

The basic idea for the scaling law for the first-order phase transition of the system with conserved particle numbers is that any quantity depending on wave number k and times $t_1, t_2, \ldots$, is scaled as

$$F_k(t_1,t_2,\ldots) = [R(t_1)]^{X_1} [R(t_2)]^{X_2} \cdots \tilde{F}(kR(t_1), kR(t_2), \ldots) \ , \tag{57}$$

where $R(t)$ is a relevant scale length at time t. The scaling law (57) is similar to those for the second-order phase transition.[33] Applying such a scaling law as (57) to the equation of motion for $S_k(t)$, which is assumed to be scaled as (1), the dependence of the scale length R on t is rederived as (2). Therefore, (57) may also be written as

$$F_k(t_1,t_2,\ldots) = t_1^{Y_1} t_2^{Y_2} \cdots \tilde{\tilde{F}}(kt_1^{1/z}, kt_2^{1/z}, \ldots) \ . \tag{57'}$$

The scaling property of the autocorrelation function $J_k(t,0)$ was discussed. Two types of the scaling law for the autocorrelation function has been discussed. One is the normal scaling law (33) or (33'), which can be naturally derived from the dynamic scaling law discussed here, and from the conservation law. The other is the anomalous scaling (35) or (35'). In the present computer simulation case (a), where $T=0.44T_c$ and $\bar{n}=0.075$, seems to follow the normal scaling (33'). Case (b), where $T=0.59T_c$ and $\bar{n}=0.075$, also seems to obey the normal scaling law for the autocorrelation function. However, we cannot deny the possibility that the autocorrelation function in this case obeys the anomalous scaling law or does not obey the scaling law. It is difficult to justify the scaling property for the autocorrelation function in this case because of the large fluctuation.

There exists a remarkable difference between the behavior of the autocorrelation functions in cases (a) and (b). Namely, the autocorrelation function in case (a) grows and then saturates, while the autocorrelation function in case (b) decays and then seems to saturate. In both cases the structure functions grow, satisfying the scaling law (1). This fact strongly suggests that there is a spinodal-like line in between two states corresponding to these cases. At this moment it is not clear which scaling law, the normal one [(33)] or the anomalous one [(35)], is usually satisfied. However, it would be correct to say that there exist two opposite behaviors in the autocorrelation functions according to whether the quenched state locates inside or outside the spinodal-like line. At this moment, however, nothing is known about the relation of the spinodal-like

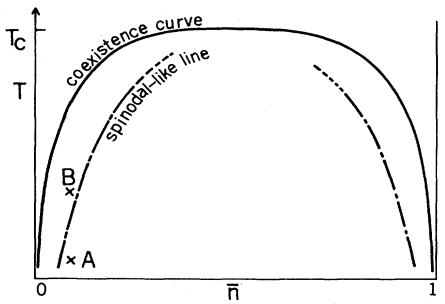

FIG. 3. Schematic aspect of the spinodal-like line. The points A and B correspond to cases (a) and (b), respectively. Inside the coexistence curve, the structure function grows. Inside the spinodal-like line the autocorrelation function would grow. In between the coexistence curve and the spinodal-like line the autocorrelation function would decay, while the structure function grows.

line identified in the present work with the spinodal line in a usual sense. The mechanisms of the phase separations in both cases are different from each other. In case (b) the growth of the structure function and therefore of the cluster size is due to the fluctuating force $f_k(t)$, while in case (a) the negative part of the damping coefficient is also important for the growth of the structure function. To find a spinodal-like line is, however, left for the future problem, together with the scaling law of the autocorrelation function in a real system.[37]

So far the discussion rests on the general consideration of dynamical scaling for the spinodal decomposition. Except for a few examples discussed in the previous paper,[8,14] the calculation of the scaling expressions for the damping coefficient Γ_k and the strength of the fluctuating force N_k has not been obtained for other cases. Various theoretical estimations of the temporal change of R, for example, the exponent z, based on the scaling idea,[8,13,23,24] however, provides us with useful information for the damping coefficient $\Gamma_k(t)$ and the strength of the fluctuating force $N_k(t)$.

We shall remark here on the essential deviation of scaling law from (1), which would be important when we calculate Γ_k and N_k by microscopic calculations. As an example, we consider the Lifshitz-Slyozov process.[23] For this process, the differences in chemical potentials among droplets are essential. Such differences in chemical potentials give only small correction to the scaling law (1). However, this correction is important to the dynamical behavior of $S_k(t)$, i.e., is effective to Γ_k and N_k. Such a small correction is also crucial to the spinodal decomposition of fluid mixture with critical concentration proposed by Siggia.[24] The chemical potential is written as

$$\mu(r)=\mu_0+\delta\mu(r) , \tag{58}$$

where μ_0 is the chemical potential at the coexistence state at a given temperature. $\delta\mu$ is a quantity of the order $1/R$ [notice that $\delta n=n-\bar{n}$ is a quantity of the order $O(1)$]. Thus, we obtain the scaling law for $\delta\mu$ as

$$\langle\mu_k(t)n_{-k}(t)\rangle_0=R^{d-1}\tilde{S}^{\mu n}(kR) , \tag{59}$$

$$\langle\mu_k(t)\mu_{-k}(t)\rangle_0=R^{d-2}\tilde{S}^{\mu\mu}(kR) . \tag{60}$$

Since the deviation of the density in a droplet from the value at the coexistence state can be regarded as a linear functional of $\delta\mu$, the deviation of the scaling law from (1) is

$$S_k(t)-R^d\tilde{S}(kR)=R^{d-1}\tilde{S}_1(kR)+O(R^{d-2}) . \tag{61}$$

Throughout this paper, we have considered the system with conserved particle numbers. A similar idea would also be possible for the system with a nonconserved order parameter.[34,38]

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