Dynamics of spinodal decomposition in finite-lifetime systems

Akira Ishikawa*
Department of Physics, Osaka University, Toyonaka, Osaka 560-0043, Japan
and Department of Physics, Tohoku University, Aoba-ku, Sendai 980-8578, Japan

Tetsuo Ogawa†
Department of Physics, Osaka University, Toyonaka, Osaka 560-0043, Japan;
Department of Physics, Tohoku University, Aoba-ku, Sendai 980-8578, Japan;
and "Structure and Transformation," PRESTO, JST, Kawaguchi 332-0012, Japan

Vladimir I. Sugakov
Institute for Nuclear Research, Ukrainian Academy of Sciences, Prospect Nauki 47, Kiev 252028, Ukraine
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We present a statistical theory of the phase-separation dynamics in many-particle systems *with a finite lifetime*. Temporal evolution of the spinodal decomposition is traced with the single-point distribution function and the dynamical structure factor under the situation where the mean particle number is constant by balancing between decay and creation of the particles. The finite lifetime prevents phase separation and order formation; hence the lower critical wave number $k_c^{(1)}(t)$ appears; domains of larger size than $[k_c^{(1)}(t)]^{-1}$ cannot grow. Differences between the infinite- and finite-lifetime cases are clarified in terms of this critical wave number. A universal relation between the lifetime and the asymptotic ($t\to\infty$) critical wave number is confirmed numerically. Comparison with the nucleation process is also made.

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I. INTRODUCTION

Spinodal decomposition is a nonequilibrium phenomenon that an unstable uniform-density state separates spatially into several stable states when a system is cooled rapidly from high temperature. This phenomenon has been studied variously since the periodic compositional variation of metallic alloys was discovered in the 1940s (see, e.g., Refs. 1–6 for reviews). In the 1960s and 1970s, it was investigated with the experiments such as x-ray scattering.[7,8] Simple linear theories were proposed by Cahn and Hilliard[9,10] and Cook,[11] which describe correctly only an early stage of the phase-separation dynamics. In the 1970s, nonlinear theories of the phase separation were developed by Langer, Bar-on, Miller[12] (LBM) and Binder.[13] They took into account not only the distribution function of the particle-number density but also the spatial correlation of the density. Binder employed Kawasaki's spin-exchange kinetic Ising model.[14] Besides these works, there are many theoretical studies on the phase separation,[15–23] the computer simulations,[24–27] and the experiments.[28–30] The phase separations are studied also in various kinds of fields such as high-polymer physics,[31] cosmology,[32] and so on.

Thus far almost all the theoretical models for the phase separation are applicable only to many-particle systems with neither creation nor annihilation of the particles. In such systems, an infinite lifetime of the particle is implicitly assumed. However, there are many examples where external fields (e.g., irradiation) create *unstable* particles (or quasiparticles in crystals) which have a *finite* lifetime and can build new phases. Several examples of such systems are listed here: (i) highly excited gas in which the excited molecules attract one another more strongly than in the ground state, leading to the creation of new phases.[33] In this case, an excited molecule is an unstable particle. (ii) The exciton-condensed liquid phase in a crystal created by light absorption,[34] in which excitons have a finite radiative lifetime. Since excitons are bosonlike quasiparticles, they are expected to occur in Bose-Einstein condensation when the excitation is stronger.[35] (iii) The electron-hole liquid in semiconductors excited more strongly than for exciton systems.[36,37] Electron-hole pairs can disappear due to recombination within a finite lifetime. It is well known that the electron-hole liquid forms a droplet.[38,39] We consider it as a phase separation of particles with finite lifetime. The phase transition between the exciton phase and the electron-hole liquid phase is also expected to occur when the density of quasiparticles is varied. This phase transition is called the exciton Mott transition.[40] (iv) New phase development in crystals under nuclear and other irradiation.[41,42] Radiation-induced microstructural space modulations were observed in many materials under various types of irradiation.[43] The phase formation occurs due to the presence of radiation defects with different types (vacancies, interstitial atoms, antisite defects, and others). Theoretical studies of the order formation such as void nucleation of vacancies, which is accompanied by the creation process and the recombination process with interstitials, were proposed.[44] In their works, however, effects of the "finite lifetime" are not noticed. The process where many vacancies aggregate to form a void is one of the phase-separation processes depending on the lifetime of the vacancies. On the other hand, the process of forming a void lattice[45] by many voids is free from the lifetime of the voids; our theory does not aim at the void lattice problem. In the above examples, the particles (or quasiparticles) have a finite value of lifetime due to light irradiation, recombination, or other processes. The finite lifetime

influences significantly the new phase development.

To the best of our knowledge, there is no theoretical framework for the phase-separation dynamics in finite-lifetime systems, where universal effects of the finite lifetime are discussed particularly. Recently, one of the authors extended the LBM theory to take into account the processes of particle creation and annihilation[46] and studied only the stationary state. In this paper, the phase-separation dynamics as a function of time is investigated to clarify effects of the finite lifetime on the phase separation.

Besides the phase separation, there are the studies in which the automatic pattern or order formation is discussed, for example, the dissipative structure,[47] synergetics,[48] and so forth.[49,50] These seem to be concerned not only with physics but also with chemistry, biology, and sociology. With respect to the appearance of stable modes in fluctuation and the creation and/or annihilation processes of components, the problem in this paper may seem to be similar to one of the dissipative structures in some chemically reacting systems.[47,51] In chemically reacting systems, however, the structures arise due to the nonlinear dependence of the creation and/or annihilation of components, whereas in our systems the structures appear due to the dynamic (attractive) interaction between particles.

This paper is organized as follows. In Sec. II, we introduce the theoretical formulation taking into account the creation and annihilation of particles with the use of the LBM approximation starting from the Ginzburg-Landau-Wilson free-energy formalism. The Lyapunov stability analysis is carried out in Sec. III to understand qualitatively the finite-lifetime effects on the phase separation. There we introduce the critical wave numbers to characterize the dynamics. Numerical results are shown in Sec. IV. We compare them with results under the simple decoupling approximation in Sec. V. The nucleation process is also studied with the same model for comparison.

II. THEORETICAL FORMULATION

A. Fokker-Planck equation for the multipoint distribution function

In the spinodal decomposition, systems exhibit spatiotemporal separation of an initial uniform-density state into two different-density states. So we define the order parameter of the system as the deviation $u(\boldsymbol{r},t)$ of the particle-number density $c(\boldsymbol{r},t)$ from its spatial average $c_0(t)\equiv\langle c\rangle(t)$, i.e., $u(\boldsymbol{r},t)\equiv c(\boldsymbol{r},t)-c_0(t)$. The state of the system is described by the multipoint distribution function $P(\{u(\boldsymbol{r})\},t)$. Using $P(\{u(\boldsymbol{r})\},t)$, the spatial average of any physical quantity $O(\{u(\boldsymbol{r})\})$ is calculated as

$$\langle O(\{u(\boldsymbol{r})\})\rangle(t)\equiv\int \mathcal{D}u(\boldsymbol{r})O(\{u(\boldsymbol{r})\})P(\{u(\boldsymbol{r})\},t), \quad (1)$$

where $\int\mathcal{D}u(\boldsymbol{r})$ is the functional integral. Temporal evolution of $P(\{u(\boldsymbol{r})\},t)$ results from two origins:

$$\frac{\partial}{\partial t}P(\{u(\boldsymbol{r})\},t)=\frac{\partial P(\{u(\boldsymbol{r})\},t)}{\partial t}\Big|_{\mathrm{c}}+\frac{\partial P(\{u(\boldsymbol{r})\},t)}{\partial t}\Big|_{\mathrm{nc}}. \quad (2)$$

The first term is the ''conservation term'' resulting from the particle transfer, in which the number of particles is conserved, and the second is the ''nonconservation term'' coming from effects of the particle creation and annihilation.

1. Conservation term: Effects of the particle transfer

We shall derive the first term resulting from the particle transfer. The free energy of the system is assumed to be the coarse-grained Ginzburg-Landau-Wilson free-energy functional

$$F[\{u(\boldsymbol{r})\}]=\int d\boldsymbol{r}\left[\frac{K}{2}(\nabla u)^2+f(u)\right], \quad (3)$$

where $f(u)$ is the Ginzburg-Landau-Wilson free-energy density:

$$f(u)=\frac{a}{2}(u+c_0-c_{\mathrm{c}})^2+\frac{b}{4}(u+c_0-c_{\mathrm{c}})^4, \quad (4)$$

and $a\equiv\tilde{a}(T-T_{\mathrm{c}})$ depends on temperature T with the critical temperature T_{c} and a real positive parameter $\tilde{a}$. Here c_{c} is the critical density, and K and b are real positive phenomenological parameters. Temporal evolution of the order parameter $u(\boldsymbol{r},t)$ caused by the particle transfer is described by the Langevin equation

$$\frac{\partial}{\partial t}u(\boldsymbol{r},t)=-\mathrm{div}\left[-M\nabla\frac{\delta F}{\delta u(\boldsymbol{r})}\right]+\eta(\boldsymbol{r},t), \quad (5)$$

where M is the mobility of the particle induced by the thermal fluctuation, $\delta/\delta u(\boldsymbol{r})$ stands for the functional derivative, and $\eta(\boldsymbol{r},t)$ is the Gaussian-Markovian noise describing a random thermal force, which satisfies

$$\langle\eta(\boldsymbol{r}_1,t_1)\eta(\boldsymbol{r}_2,t_2)\rangle=-2k_BTM\nabla^2\delta(\boldsymbol{r}_1-\boldsymbol{r}_2)\delta(t_1-t_2). \quad (6)$$

We rewrite the Langevin equation (5) into the Fokker-Planck equation for the multipoint distribution function as

$$\frac{\partial P(\{u(\boldsymbol{r})\},t)}{\partial t}\Big|_{\mathrm{c}}=-\int d\boldsymbol{r}\frac{\delta}{\delta u(\boldsymbol{r})}\left(M\nabla^2\left[\frac{\delta F}{\delta u(\boldsymbol{r})}P(\{u(\boldsymbol{r})\},t)+k_BT\frac{\delta}{\delta u(\boldsymbol{r})}P(\{u(\boldsymbol{r})\},t)\right]\right). \quad (7)$$

This is the conservation term of Eq. (2).

2. Nonconservation term: Effects of the particle creation and annihilation

The creation rate of particles is defined as $y(t)$, which is a given function of t. Here the particle creation is assumed to be homogeneous in space; hence $y(t)$ is independent of the position $\boldsymbol{r}$. The particle lifetime is denoted as τ, which depends on neither t nor $\boldsymbol{r}$. This assumption for the lifetime is the simplest as the first step for future studies of the phase-separation dynamics in finite-lifetime systems. Extension beyond this assumption is straightforward.[52] In order to introduce the particle creation and annihilation processes, we shall divide the space into $N_l(\gg 1)$ pieces of small regions

$\{\Delta_l | l=1,\ldots,N_l\}$. The number of particles in the lth small region Δ_l is denoted as n_l. Temporal evolution of the distribution function $P(\{n_l\},t)$ for the particle numbers $\{n_l\}$ in all small regions $\{\Delta_l\}$ obeys the Master equation[46]

$$\frac{\partial}{\partial t}P(\{n_l\},t)|_{\rm nc}=\sum_l \Big[y(t)\Delta_l P(\{n_1,\ldots,n_l-1,\ldots,n_{N_l}\},t) - y(t)\Delta_l P(\{n_l\},t) + \frac{n_l+1}{\tau}P(\{n_1,\ldots,n_l+1,\ldots,n_{N_l}\},t) - \frac{n_l}{\tau}P(\{n_l\},t)\Big], \tag{8}$$

where $P(\{n_1,\ldots,n_l\pm 1,\ldots,n_{N_l}\},t)$ is the probability distribution function when the particle configuration is $\{n_1,\ldots,n_l\pm 1,\ldots,n_{N_l}\}$. The summation Σ_l runs over all N_l pieces of small regions $\{\Delta_l\}$.

To derive the Fokker-Planck equation for $P(\{u(\boldsymbol{r})\},t)$, a coarse-graining procedure in a small region is required. First, we assume that $n_l \gg 1$ and that n_l is continuous quantity. Then $P(\{n_1,\ldots,n_l\pm 1,\ldots,n_{N_l}\},t)$ can be expanded as

$$P(\{n_1,\ldots,n_l\pm 1,\ldots,n_{N_l}\},t) = P(\{n_l\},t)\pm\frac{\partial}{\partial n_l}P(\{n_l\},t)+\frac{1}{2}\frac{\partial^2}{\partial n_l^2}P(\{n_l\},t). \tag{9}$$

Second, we rewrite n_l into u_l as $u_l \equiv n_l/\Delta_l - c_0(t)$, where u_l is the order parameter in the lth small region Δ_l. Finally, we take the limit of $\Delta_l \to 0$, in which the spatial coordinate becomes continuous. Then the summation Σ_l over all small regions is rewritten as the space integral $\int d\boldsymbol{r}$.

Performing these transformations, we get the Fokker-Planck equation for $P(\{u(\boldsymbol{r})\},t)$ including effects of the particle creation and annihilation:

$$\frac{\partial P(\{u(\boldsymbol{r})\},t)}{\partial t}|_{\rm nc}=-\int d\boldsymbol{r}\frac{\delta}{\delta u(\boldsymbol{r})}\Bigg(\Big[y(t)-\frac{u(\boldsymbol{r})+c_0(t)}{\tau}\Big]\times P(\{u(\boldsymbol{r})\},t) + \frac{1}{2}\frac{\delta}{\delta u(\boldsymbol{r})}\Big[\Big\{y(t)+\frac{u(\boldsymbol{r})+c_0(t)}{\tau}\Big\}\times P(\{u(\boldsymbol{r})\},t)\Big]\Bigg). \tag{10}$$

This is the nonconservation term of Eq. (2). Here the mean particle density $c_0(t)$ depends on time due to creation and annihilation of particles. Using Eqs. (2), (7), and (10), we derived the full form of the Fokker-Planck equation for the multipoint distribution function $P(\{u(\boldsymbol{r})\},t)$ in a system with the particle creation and annihilation effects.

B. Closed-form equations of motion: The LBM approximation

The Fokker-Planck equations, (2), (7), and (10) for the multipoint distribution function contains all the statistical, spatiotemporal information of the phase-separation dynamics. Unfortunately, however, it is hard to solve. Then we shall divide again continuous space into discrete $N_0(\gg 1)$ cells (with an index α) to discard the higher-order statistical information in the multipoint distribution function. In a cell, the particle density is assumed to be constant, i.e., $u(\boldsymbol{r})|_{\boldsymbol{r}=\boldsymbol{r}_\alpha \in \alpha\text{th cell}} \equiv u_\alpha$. In our formulation, the state of the system is described with two quantities. One is the single-point distribution function

$$P^{(1)}(u_\alpha,t)\equiv\int\prod_{m\neq\alpha}du_m P(\{u_m\},t). \tag{11}$$

This is the probability distribution for the case that the order parameter in the αth cell takes the value u_α at time t. Since α is arbitrary ($\alpha=1,2,\ldots,N_0$), $P^{(1)}(u_\alpha,t)$ means the distribution function of the number of cells in which the particle density is $c_\alpha=u_\alpha+c_0$. In the cell picture, an averaged physical quantity $O(\{u_\alpha\})$ is given as

$$\langle O(\{u_\alpha\})\rangle(t)\equiv\int\prod_\alpha du_\alpha O(\{u_\alpha\})P(\{u_\alpha\},t), \tag{12}$$

where the functional integral $\int\mathcal{D}u(\boldsymbol{r})$ is transformed to $\int\Pi_\alpha du_\alpha$. If the quantity $O(u_\alpha)$ depends only on u_α (not on u_β), the averaging is carried out with only $P^{(1)}(u_\alpha,t)$,

$$\langle O(u_\alpha)\rangle(t)=\int du_\alpha O(u_\alpha)P^{(1)}(u_\alpha,t). \tag{13}$$

Another quantity for describing the state is the dynamical structure factor

$$S(\boldsymbol{k},t)=\sum_\alpha \exp[-i\boldsymbol{k}\cdot(\boldsymbol{r}_\alpha-\boldsymbol{r}_\beta)]\langle u_\alpha u_\beta\rangle(t), \tag{14}$$

where $\boldsymbol{k}$ is the wave vector. Here $\langle u_\alpha u_\beta\rangle(t)$ is the two-point spatial correlation function at time t, which is a function of $\boldsymbol{r}_\alpha-\boldsymbol{r}_\beta$. This structure factor corresponds approximately to the distribution of the domain size $|\boldsymbol{k}|^{-1}$ (i.e., a spatial correlation length), which can be measured by actual experiments such as x-ray or light scattering.

In order to derive a closed form of coupled equations for the single-point distribution function $P^{(1)}(u,t)$ and the dynamical structure factor $S(\boldsymbol{k},t)$ from Eqs. (2), (7), and (10), we need to truncate the hierarchy of the time-evolution equations for the probability distribution functions. To this end, we employ an approximation[12] in which the two-point distribution function is decoupled as

$$P^{(2)}(u_\alpha,u_\beta,t)=P^{(1)}(u_\alpha,t)P^{(1)}(u_\beta,t)\times\Big[1+\frac{\langle u_\alpha u_\beta\rangle(t)}{[\langle u^2\rangle(t)]^2}u_\alpha u_\beta\Big], \tag{15}$$

where $\langle u^2\rangle(t)$ is the variance of u at time t, and α and β stand for two different cells. In this paper, we call this approximation the ''LBM approximation.'' With the use of the LBM approximation, the closed-form coupled equations for $P^{(1)}(u,t)$ and $S(\boldsymbol{k},t)$ are derived. But several phenomenological parameters still remain in such equations. To eliminate them, physical quantities are renormalized by the following units: $u_0 \equiv (-a/b)^{1/2}$ for density, $(-K/a)^{1/2}$ for length, $(-K^3a/b^2)^{1/2}$ for energy, and $K/(Ma^2)$ for time. The unit density u_0 is concerned with the equilibrium values of the order parameter, $c_c - c_0 \pm u_0$, in the case of neither creation nor annihilation of particles, i.e., $y(t) \equiv 0$ and $\tau \to \infty$. To discuss the spinodal decomposition, the temperature T is chosen to be lower than the critical temperature T_c; that is, we confine ourselves to the case of $a<0$. The Landau free-energy density (4) can be written in a dimensionless form as $f(u) = -\frac{1}{2}(u+c_0-c_c)^2 + \frac{1}{4}(u+c_0-c_c)^4$, where c_0 and c_c are normalized by the unit u_0. Accordingly, the renormalized order parameter becomes independent of the index α in our model. For the equilibrium stable state, the normalized order parameter is $c_c - c_0 \pm 1$.

Applying the LBM approximation and the above-mentioned normalization, the closed-form coupled equations for $P^{(1)}(u,t)$ and $S(\boldsymbol{k},t)$ are obtained as

$$\frac{\partial}{\partial t}P^{(1)}(u,t) = \frac{\partial}{\partial u}\left\{\left[D_f\left(\frac{\partial f}{\partial u} - \left\langle\frac{\partial f}{\partial u}\right\rangle(t) - A(t)u\right) + \frac{W(t)}{\langle u^2\rangle(t)}u - \left(y(t) - \frac{u+c_0(t)}{\tau}\right)\right]P^{(1)}(u,t) + D_f T\frac{\partial}{\partial u}P^{(1)}(u,t) + \frac{1}{2v_0u_0}\frac{\partial}{\partial u} \times \left(\left[y(t) + \frac{u+c_0(t)}{\tau}\right]P^{(1)}(u,t)\right)\right\}, \tag{16}$$

$$\frac{\partial}{\partial t}S(\boldsymbol{k},t) = -2\left[k^2\{k^2 + A(t)\} + \frac{1}{\tau}\right]S(\boldsymbol{k},t) + 2Tk^2 + \frac{1}{v_0u_0}\left[y(t) + \frac{c_0(t)}{\tau}\right], \tag{17}$$

where v_0 is the volume of a cell and

$$A(t) \equiv \left(\frac{\partial^2 f}{\partial u^2}\right)_{u=0} + \frac{1}{2}\left[\left(\frac{\partial^3 f}{\partial u^3}\right)_{u=0}\right]\frac{\langle u^3\rangle(t)}{\langle u^2\rangle(t)} + \frac{1}{6}\left[\left(\frac{\partial^4 f}{\partial u^4}\right)_{u=0}\right]\frac{\langle u^4\rangle(t)}{\langle u^2\rangle(t)}, \tag{18}$$

$$W(t) \equiv \frac{1}{(2\pi)^3}\int d\boldsymbol{k}k^2\{k^2 + A(t)\}S(\boldsymbol{k},t). \tag{19}$$

Here the $\boldsymbol{k}$ integration is carried out in the range of $0 \leqslant |\boldsymbol{k}| \leqslant v_0^{-1/3}$ and $-D_f$ is the diagonal element of the renormalized Laplacian matrix defined as

$$-D_f = -\frac{1}{(2\pi)^3}\int d\boldsymbol{k}k^2. \tag{20}$$

The mean value is obtained by Eq. (13). The mean particle-number density $c_0(t)$ depends on time as

$$\frac{dc_0(t)}{dt} = y(t) - \frac{c_0(t)}{\tau}. \tag{21}$$

In this paper, we shall investigate only the case where the particles are created at a constant rate (''cw creation'') to balance with the depletion of particles due to the finite lifetime; that is, both y and the mean particle-number density $c_0 = y\tau$ are constant in time. Such a situation is realized, e.g., in an exciton system. We keep the temperature at $T > T_c$ under cw irradiation and then decrease abruptly the temperature to $T < T_c$ at a time. The reasons why we consider such a restrictive situation as cw creation are that the case of the finite lifetime can be directly compared with the infinite-lifetime case (i.e., the usual spinodal decomposition) and that effects only of the ''finite lifetime'' can be picked out easily. When more realistic problems are discussed, we can straightforwardly calculate various situations with the same formulation in this paper.

C. Comments on another approximation scheme

We here make comments on another approximation for truncation of the hierarchy of the equations of motion. In the LBM approximation, information of the two-point spatial correlation function $\langle u_\alpha u_\beta\rangle(t)$ is fully taken into account in the decoupling procedure from $P^{(2)}(u_\alpha,u_\beta,t)$ to $P^{(1)}(u_\alpha,t)$ and $P^{(1)}(u_\beta,t)$, as shown in Eq. (15). For example, a higher-order nonlinear term $\langle u_\alpha^3 u_\beta\rangle(t)$ is transformed with the use of the LBM approximation as

$$\langle u_\alpha^3 u_\beta\rangle(t) \to \frac{\langle u^4\rangle(t)}{\langle u^2\rangle(t)}\langle u_\alpha u_\beta\rangle(t). \tag{22}$$

Equations (16) and (17) contain such nonlinear terms as $A(t)$ of Eq. (18).

There are other methods for approximating the nonlinear terms such as $\langle u_\alpha^3 u_\beta\rangle(t)$. One method is the so-called ''decoupling approximation''

$$\langle u_\alpha^3 u_\beta\rangle(t) \to \langle u^2\rangle(t)\langle u_\alpha u_\beta\rangle(t). \tag{23}$$

If we employ this decoupling approximation, the nonlinear term $A(t)$ might be

$$A_{\text{decouple}}(t) = \left(\frac{\partial^2 f}{\partial u^2}\right)_{u=0} + \frac{1}{6}\left[\left(\frac{\partial^4 f}{\partial u^4}\right)_{u=0}\right]\langle u^2\rangle(t). \tag{24}$$

Since the time-dependent variance $\langle u^2\rangle(t)$ in Eq. (24) can be calculated only from the dynamical structure factor $S(\boldsymbol{k},t)$ as

$$\langle u^2\rangle(t) = \frac{1}{(2\pi)^3}\int d\boldsymbol{k}S(\boldsymbol{k},t), \tag{25}$$

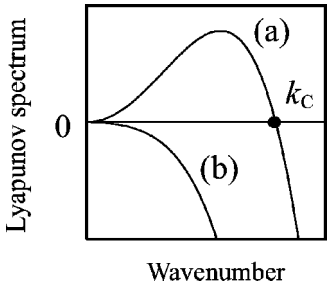

FIG. 1. The Lyapunov spectrum $\Gamma_k(t)$ with neither creation $[y(t)\equiv 0]$ nor annihilation $(\tau\to\infty)$ of particles as a function of wave number k when (a) $A(t)<0$ and (b) $A(t)>0$.

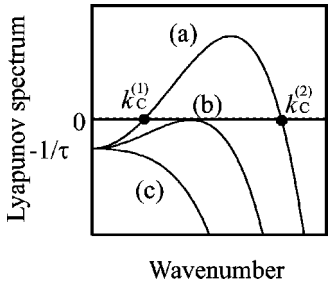

FIG. 2. The Lyapunov spectrum $\Gamma_k(t)$ with cw creation and annihilation $(y\tau=\text{const}<\infty)$ of particles as a function of wave number k when (a) $A(t)<-2/\sqrt{\tau}$, (b) $A(t)=-2/\sqrt{\tau}$, and (c) $A(t)>-2/\sqrt{\tau}$.

the equation of motion consist only of Eq. (17) for the dynamical structure factor. Thus far, this decoupling approximation has often been employed and the temporal evolution only of the dynamical structure factor has been calculated. It is clear that this approximation is worse than the LBM one because it does not contain higher-order statistical information such as $\langle u^3\rangle(t)$ or $\langle u^4\rangle(t)$, which are taken in the LBM formalism. In addition, the LBM approximation is more favorable for our study since we can obtain also temporal information of the single-point distribution function. We will discuss later the numerical results in more detail.

III. LYAPUNOV STABILITY ANALYSIS

Before we discuss in detail the numerical results, the Lyapunov stability analysis is carried out to understand qualitatively the dynamics. The temporal behavior of the dynamical structure factor is roughly understood by a few characteristic wave numbers. To obtain them, we assume the density-fluctuation mode $u_{\boldsymbol{k}}(t)$ with wave number $\boldsymbol{k}$ of the form

$$u_{\boldsymbol{k}}(t)\propto\exp[t\Gamma_{\boldsymbol{k}}(t)]. \tag{26}$$

Here $\Gamma_{\boldsymbol{k}}(t)$ is called the ''Lyapunov spectrum,'' which means the growing speed of a fluctuation mode of wave number $\boldsymbol{k}$. The fluctuation mode grows if $\Gamma_{\boldsymbol{k}}(t)>0$ and decays if $\Gamma_{\boldsymbol{k}}(t)<0$. The Lyapunov spectrum describes also the increase or decrease of the dynamical structure factor through the relation $S(\boldsymbol{k},t)=|u_{\boldsymbol{k}}(t)|^2\propto\exp[2t\Gamma_{\boldsymbol{k}}(t)]$. Thus the Lyapunov spectrum is a good measure for the growing speed of domains with size $|\boldsymbol{k}|^{-1}$. In our formulation, systems are assumed to be isotropic. Therefore the wave vector $\boldsymbol{k}$ can be replaced simply by its amplitude $k\equiv|\boldsymbol{k}|$.

In the case of $y(t)=c_0/\tau=\text{const}$, the Lyapunov spectrum is

$$\Gamma_k(t)=-k^2[k^2+A(t)]-\frac{1}{\tau}. \tag{27}$$

The calculated Lyapunov spectra at time t are shown in Figs. 1 and 2 as a function of the wave number k. In the figures, we shall define the critical wave number $k_c(t)$ as a zero point of $\Gamma_k(t)$ at a fixed time t, which is the wave number at a boundary between unstable $[\Gamma(k,t)>0]$ and stable $[\Gamma(k,t)<0]$ density-fluctuation modes.

In systems with neither creation nor annihilation of particles $[y(t)\equiv 0$ and $\tau\to\infty]$, it is clear from Fig. 1 that there is only *one* critical wave number $k_c(t)$, which is $k_c(t)=\sqrt{-A(t)}$. This is consistent with the well-known process of spinodal decomposition. Domains with larger size $(>[k_c(t)]^{-1})$ grow but domains with smaller size $(<[k_c(t)]^{-1})$ decay. In the case of $y(t)=c_0/\tau=\text{const}$, on the other hand, we find from Fig. 2 that there are *two* critical wave numbers, the lower critical wave number $k_c^{(1)}(t)$ and the upper critical wave number $k_c^{(2)}(t)$, which are evaluated as

$$k_c^{(1)}(t)=\left(\frac{-A(t)-\sqrt{[A(t)]^2-4/\tau}}{2}\right)^{1/2}, \tag{28}$$

$$k_c^{(2)}(t)=\left(\frac{-A(t)+\sqrt{[A(t)]^2-4/\tau}}{2}\right)^{1/2}. \tag{29}$$

This means that the density-fluctuation mode only of wave number $k_c^{(1)}(t)<k<k_c^{(2)}(t)$ can grow and the mode of smaller wave number $k<k_c^{(1)}(t)$ decays in time. In other words, domains with very large size $(>[k_c^{(1)}(t)]^{-1})$ cannot grow in the case of $y(t)=c_0/\tau=\text{const}$, which results from the finite-lifetime effect. This is in striking contrast to the case of $y(t)\equiv 0$ and $\tau\to\infty$, where $k_c^{(1)}$ vanishes. In Sec. IV, the temporal behavior of the critical wave numbers will be discussed numerically.

IV. NUMERICAL RESULTS AND DISCUSSION

The coupled equations (16) and (17) are solved numerically without assuming the double-Gaussian ansatz used by LBM.[12] In calculations, we set the mean particle-number density to be $c_0=c_c=1.5=\text{const}$ in the spinodal region. The lifetime is chosen to be $\tau=\infty$, 100, and 8. The cw creation rate y satisfies the relation $y=c_0/\tau=1.5/\tau$. The temperature is always lower than the critical temperature T_c, i.e., $T=0.1$. The parameter v_0u_0, which is almost equal to the number of particles in one cell, is taken to be $v_0u_0=1000$, which should not be so small. Final numerical results are insensitive to this value v_0u_0 if it is large enough. The initial state at $t=0$ is chosen to be a uniform-density state. Then the initial single-point distribution function has a δ-function-like form with a small width due to the thermal fluctuation. In

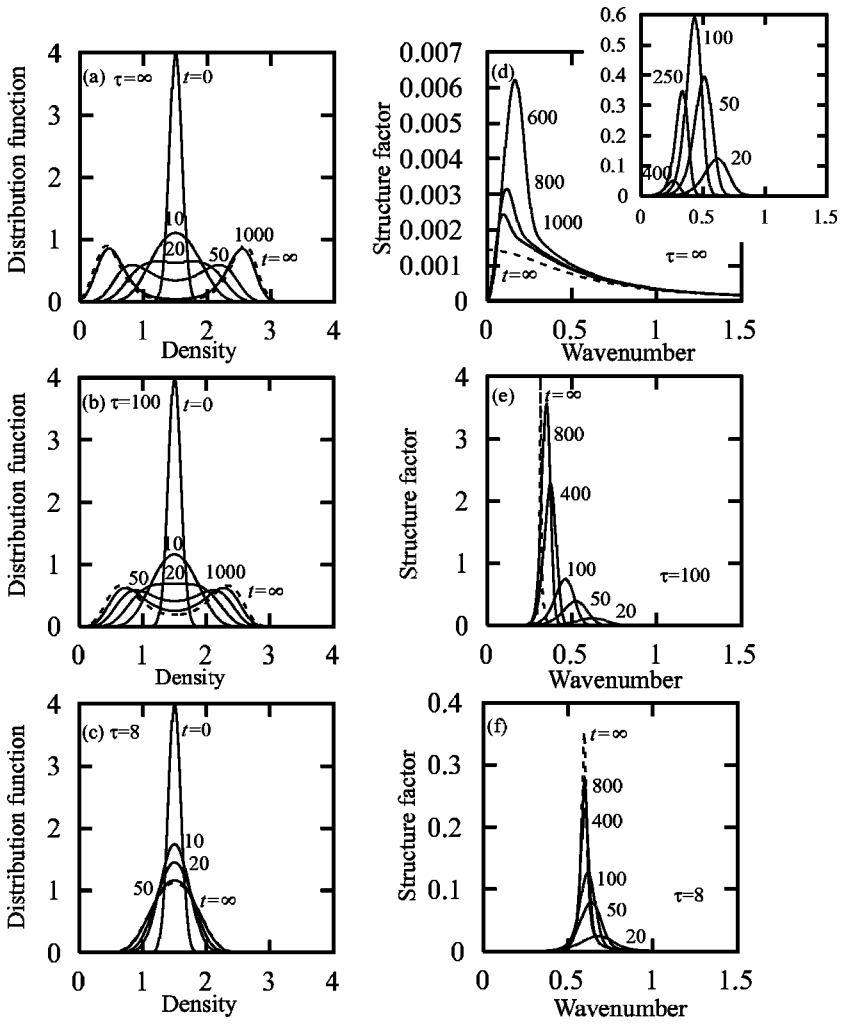

FIG. 3. Temporal development of the single-point distribution function $P^{(1)}(u,t)$ and the dynamical structure factor $S(k,t)$ as a function of u and k, respectively, for $T=0.1$, $v_0u_0=1000$, and $c_0=c_c=1.5$. The creation rate is $y=c_0/\tau=1.5/\tau$. The three figures in the left row, (a), (b), and (c), are $P^{(1)}(u,t)$ and the ones in the right row, (d), (e), and (f), are $S(k,t)$. The lifetime of particles is $\tau=\infty$ for (a) and (d), 100 for (b) and (e), and 8 for (c) and (f). The inset of (d) shows the behavior during $0\leqslant t\lesssim 400$. The dashed curves stand for the stationary solutions at $t\to\infty$.

numerical calculations, we assume it as a Gaussian, whose standard deviation is 0.1. The boundary condition for the single-point distribution function is $P^{(1)}(u,t)|_{u<-c_0}=0$, which results from the fact that the particle-number density should be positive. The initial dynamical structure factor is assumed to be $S(k,t=0)=0$, which reflects that no domain structure exists at $t=0$. According to the coarse-graining procedure for the cell representation in Sec. II B, the largest normalized wave number should be $(6\pi^2)^{1/3}$, which is derived from the fact that the number of states is equal to the number of cells.

A. Single-point distribution function and dynamical structure factor

The numerical results are shown in Fig. 3. Figures 3(a) and 3(d) correspond to the case of neither creation nor annihilation of particles and Figs. 3(b), 3(c), 3(e), and 3(f) to the case where particle creation and annihilation are taken into account. The dashed lines mean the stationary solutions at $t\to\infty$.

First, we discuss the behavior of the single-point distribution function. In the case of $\tau=\infty$ [correspondingly $y(t)\equiv 0$], as shown in Fig. 3(a), the initial state with one uniform density separates into two states with different densities. This is nothing but the usual spinodal decomposition. Here we shall define the ''onset time'' $t_{\rm onset}$ as the time when one peak of $P^{(1)}(u,t)$ is about to separate into two peaks. In the case of Fig. 3(a), the onset time is $t_{\rm onset}\simeq 20$. In the case of a finite lifetime $\tau=100$, which is longer than the onset time, on the other hand, the phase separation of the order parameter becomes ambiguous as shown in Fig. 3(b). When the lifetime $\tau=8$ is much shorter than the onset time in Fig. 3(c), phase separation cannot occur. Consequently, order formation or phase separation stops halfway due to finite-lifetime effects if the lifetime is shorter than the onset time, which agrees with intuition.

Next, we discuss the behavior of the dynamical structure factor. Here we denote $k_{\rm m}(t)$ as the wave number at which the dynamical structure factor takes the maximum value. In the case of $\tau=\infty$ [correspondingly $y(t)\equiv 0$], as shown in Fig. 3(d), the dynamical structure factor grows until $t\lesssim 100$ but declines later, as shown in the inset of Fig. 3(d). When $t\to\infty$, the dynamical structure factor becomes a form plotted as a dashed line in Fig. 3(d), which is a Lorentzian with a peak at $k=0$. We interpret this growing and declining of the dynamical structure factor as follows. A schematic behavior of the order parameter on the Landau free-energy density is shown in Fig. 4. The initial state is plotted as an open circle.

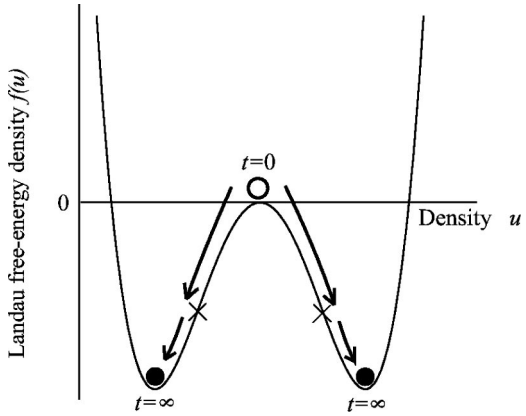

FIG. 4. Temporal development of the order parameter u is schematically drawn along the Landau free-energy density curve when neither creation nor annihilation of particles exists. The open circle stands for the initial state. The order parameter separates into two directions as time passes. After the cross points, where $d^2f(u)/du^2=0$ holds, the system goes to the stable regime. Finally $(t\rightarrow\infty)$, the system reaches an equilibrium stable state and the order parameter gets to the solid-circle points.

At this time, the system is unstable, so the unstable mode grows. Due to the increase of the instability, the dynamical structure factor grows as a whole. The order parameter separates into two directions as time passes. Two cross symbols indicate the boundary points between the unstable and stable regimes, at which $d^2f(u)/du^2=0$ holds. After the order parameters pass through these points, the system tends to be stable. So the smaller unstable domains vanish and the larger stable domains survive and increase. Accordingly, the dynamical structure factor declines as a whole. It is expected that the time when the order parameters pass through the cross points is around $t\simeq 100$. The wave number $k_{\mathrm{m}}(t)$ shifts to the longer-wavelength side as shown in Fig. 3(d). Finally, when $t\rightarrow\infty$, it is found that $k_{\mathrm{m}}(t\rightarrow\infty)=0$, where the system separates completely into two phases.

The cases of the finite lifetime $\tau=100$ and 8 are shown in Figs. 3(e) and 3(f), respectively, where the temporal development of $S(k,t)$ is quite different from the infinite-lifetime case in Fig. 3(d). The form becomes sharp and $k_{\mathrm{m}}(t)$ settles into a finite wave number as $t\rightarrow\infty$. This fact is interpreted as that the system cannot separate completely and that the finite-size domains remain finally. Comparing Figs. 3(e) with 3(f), the shorter the lifetime of the particles is, the larger $k_{\mathrm{m}}(t\rightarrow\infty)$ becomes. We find from the numerical results that there is a universal relationship between $k_{\mathrm{m}}(t\rightarrow\infty)$ and the lifetime τ:

$$k_{\mathrm{m}}(t\rightarrow\infty)\propto\tau^{-1/4}. \tag{30}$$

The final domain size $(\propto[k_{\mathrm{m}}(t\rightarrow\infty)]^{-1})$ is determined by the competition between the lifetime and the time necessary for gathering the particles. If the lifetime is short enough, the particles decay before they come together. So the final domain size becomes smaller. The above relation shows this fact.

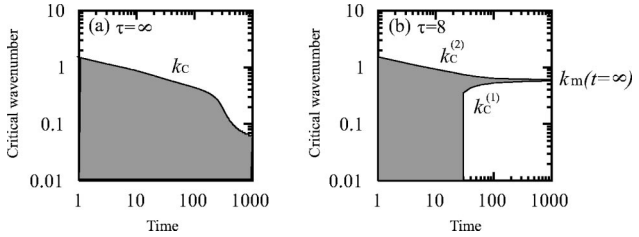

FIG. 5. Temporal development of the critical wave numbers for (a) $\tau=\infty$ and (b) $\tau=8$. In (a) there is only one critical wave number $k_{\mathrm{c}}(t)$, while there are two critical wave numbers $k_{\mathrm{c}}^{(1)}(t)$ and $k_{\mathrm{c}}^{(2)}(t)$ in (b). The shadowed regions indicate the *unstable* density-fluctuation modes. At $t\rightarrow\infty$, $k_{\mathrm{c}}(\infty)=0$ in (a), while $k_{\mathrm{c}}^{(1)}(\infty)=k_{\mathrm{c}}^{(2)}(\infty)=k_{\mathrm{m}}(\infty)>0$ in (b).

B. Critical wave numbers

Figure 5 shows the behavior of the critical wave numbers for (a) $\tau=\infty$ and (b) $\tau=8$. In the case of $\tau=\infty$, there is only one critical wave number $k_{\mathrm{c}}(t)$, as explained in Sec. III. The density-fluctuation modes in a shadowed region $k<k_{\mathrm{c}}(t)$ are unstable and grow in time. We find that the abrupt change of the critical wave number $k_{\mathrm{c}}(t)$ takes place at $t\simeq 200$. This step may result from a crossover between two different types of domain growth: from droplet formation to coalescence.

In the case of $\tau=8$, on the other hand, there are two kinds of critical wave numbers: the lower critical wave number $k_{\mathrm{c}}^{(1)}(t)$ and the upper critical wave number $k_{\mathrm{c}}^{(2)}(t)$, as explained in Sec. III. The main difference from the $\tau=\infty$ case is that the lower critical wave number $k_{\mathrm{c}}^{(1)}(t)$ appears and that growth of the modes in the region of $k<k_{\mathrm{c}}^{(1)}(t)$ is restrained, as shown in Fig. 5(b) for $\tau=8$. The modes only in the shadowed region $k_{\mathrm{c}}^{(1)}(t)<k<k_{\mathrm{c}}^{(2)}(t)$ can grow. It is found that the shadowed region $k_{\mathrm{c}}^{(1)}(t)<k<k_{\mathrm{c}}^{(2)}(t)$ becomes narrow and finally remains only at a particular finite wave number, i.e., $k_{\mathrm{c}}^{(1)}(t\rightarrow\infty)=k_{\mathrm{c}}^{(2)}(t\rightarrow\infty)=k_{\mathrm{m}}(t\rightarrow\infty)$. This means that size of all the domains is finite and determined by the competition between the lifetime and the time necessary for the particles' gathering. This is in striking contrast to the case of $\tau=\infty$.

V. DISCUSSION AND CONCLUSIONS

A. Comparison with the decoupling approximation

We examine the numerical results under the decoupling approximation. To discuss differences coming only from the approximation methods, we confine ourselves to the case of $\tau=\infty$ [accordingly $y(t)\equiv 0$]. Figure 6(a) is the temporal evolution of the dynamical structure factor and Fig. 6(b) is the critical wave number $k_{\mathrm{c}}(t)$ under the decoupling approximation. Under the decoupling approximation, as we mentioned, we cannot calculate the single-point distribution function. The parameters in Fig. 6 are the same as in Figs. 3(a) and 3(d). We shall compare Fig. 6(a) with Fig. 3(d). In the case of the LBM approximation, the dynamical structure factor grows first and declines later. In the case of the decoupling approximation, on the other hand, it goes on growing and does not decline later. Next, let us compare Fig. 6(b) with Fig. 5(a). In the case of the decoupling approximation, the

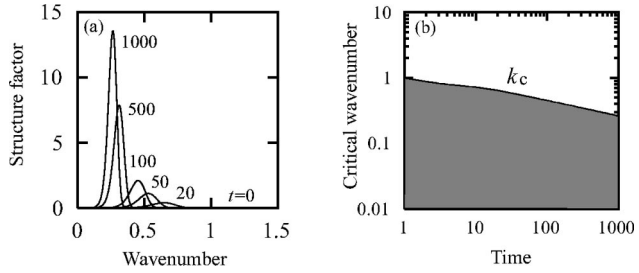

FIG. 6. Numerical results of (a) the dynamical structure factor and (b) the critical wave number with the use of the decoupling approximation in the case of $\tau\to\infty$ and $y(t)\equiv 0$. Parameters are $T=0.1$, $v_0u_0=1000$, and $c_0=c_c=1.5$. The dynamical structure factor moves to the longer-wavelength side and always grows. The critical wave number $k_c(t)$ decreases monotonously.

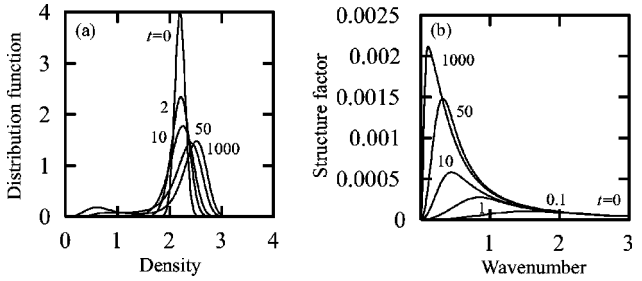

FIG. 7. Temporal development of the single-point distribution function $P^{(1)}(u,t)$ and the dynamical structure factor $S(k,t)$ as a function of u and k, respectively, for the nucleation process in the case of $\tau\to\infty$ and $y(t)\equiv 0$. Parameters are $T=0.1$, $v_0u_0=1000$, $c_c=1.5$, and $c_0=2.2$.

sudden change of the critical wave number disappears, which contrasts to the case of the LBM approximation, where an abrupt change takes place.

As was mentioned, the alteration from growing to declining of the dynamical structure factor and the abrupt change of the critical wave number are caused by crossover from the unstable regime to the stable one of the system. However, the decoupling approximation is valid only when the distribution function has a Gaussian form, which cannot describe a phase-separated state with the two-peak distribution function. Therefore the decoupling approximation is invalid for this crossover phenomenon. This is a reason why the characteristic behavior of $S(k,t)$ and $k_c(t)$ under the LBM approximation cannot be reproduced with the decoupling approximation. Thus we can conclude that the LBM approximation is more appropriate than the decoupling approximation in the study of the phase-separation dynamics.

B. Comparison with the nucleation process

We shall consider also the nucleation dynamics to compare with the spinodal decomposition in the case of infinite lifetime $\tau=\infty$ in terms of $P^{(1)}(u,t)$ and $S(k,t)$. Before discussing numerical details, we here explain a qualitative difference between the two processes. To this end, the Lyapunov spectrum under the crude linear approximation is employed:

$$\Gamma_k=-k^2\left[k^2+\left(\frac{\partial^2 f}{\partial u^2}\right)_{u=0}\right]. \tag{31}$$

The linear approximation, which excludes all nonlinear terms depending on time, is valid only at the early stage of the phase-separation dynamics. The boundary between the spinodal decomposition and the nucleation process is defined through the relation

$$\left(\frac{\partial^2 f}{\partial u^2}\right)_{u=0}=3(c_0-c_c)^2-1=0. \tag{32}$$

If the initial mean density $c_0(t=0)$ is in the region of

$$c_c-\frac{1}{\sqrt{3}}<c_0(t=0)<c_c+\frac{1}{\sqrt{3}}, \tag{33}$$

the wave numbers satisfying $\Gamma_k>0$ can exist for $k<-(|\partial^2 f/\partial u^2|_{u=0})^{1/2}$, in which the density-fluctuation modes are unstable and grow in time. This corresponds to the spinodal decomposition at an early stage. On the other hand, if the initial mean density $c_0(t=0)$ is in the region of

$$c_0(t=0)<c_c-\frac{1}{\sqrt{3}}\ \text{ or }\ c_c+\frac{1}{\sqrt{3}}<c_0(t=0), \tag{34}$$

no wave number satisfying $\Gamma_k>0$ exists. In this case, all density-fluctuation modes are stable. This corresponds to the nucleation dynamics, although, of course, small droplets are always created due to the thermal fluctuations. We calculate $P^{(1)}(u,t)$ and $S(k,t)$ for the nucleation regime in the case of infinite lifetime $\tau=\infty$.

Figure 7 shows the temporal evolution of $P^{(1)}(u,t)$ and $S(k,t)$ for the mean density $c_0=2.2$ and the critical density $c_c=1.5$. The other parameters are the same as in Figs. 3(a) and 3(d) for the spinodal decomposition. Because the mean density c_0 is larger than the critical density c_c, the single-point distribution function is asymmetrical, as shown in Fig. 7(a). The width of $P^{(1)}(u,t)$ is broad and the peak shifts to the larger-density side. As the time goes by, $P^{(1)}(u,t)$ oozes to the smaller-density side and another small peak appears. A clearer difference between the spinodal decomposition [Fig. 3(d)] and the nucleation process [Fig. 7(b)] is found in terms of the dynamical structure factor $S(k,t)$. Recall that $S(k,t)$ grows first and then declines later in the case of the spinodal decomposition, as shown in Fig. 3(d). In the case of the nucleation process, on the other hand, $S(k,t)$ always grows, as shown in Fig. 7(b).

C. Conclusions

We have formulated a theoretical model to discuss the dynamical characteristics of the phase separation in finite-lifetime systems with the LBM approximation. The temporal behavior of the single-point distribution function $P^{(1)}(u,t)$ and the dynamical structure factor $S(k,t)$ has been calculated numerically in the case of cw creation of particles. We can

calculate straightforwardly the other cases, where the creation rate depends on time. When the lifetime is enough short $P^{(1)}(u,t)$ cannot separate into two peaks and $S(k,t)$ becomes a sharp-peak form at a finite wave number. These peculiar characteristics in the finite-lifetime case are well understood in terms of the lower and upper critical wave numbers $k_c^{(1)}(t)$ and $k_c^{(2)}(t)$, respectively. Domains with a finite size of $[k_c^{(1)}(t\to\infty)]^{-1}$ remain finally, the size of which is proportional to $\tau^{1/4}$. The differences between the LBM approximation and the decoupling approximation were clarified in the case of infinite lifetime $\tau=\infty$. The former is better for the study of the phase-separation dynamics. We have compared also the spinodal decomposition with the nucleation process in the case of infinite lifetime.

As a final discussion in this paper, we describe a few things about the extension of this theory. It is clarified that the phase separation is incomplete when the lifetime is shorter than the onset time. In order to obtain the onset time we need to discuss more microscopic characteristics of the system such as the interaction between particles, which is implicitly built in the phenomenological Ginzburg-Landau-Wilson free-energy functional in this paper. Theoretical study of this viewpoint is in progress.[53] As another extension, we are interested also in the case where the lifetime depends on the local density of particles, in other words, where the annihilation term is nonlinearly dependent on the particle density. For example, a system with the two-particle collision process is investigated.[52] In this paper, we have considered only the simplest case as the first step in investigations for finite-lifetime effects. An actual situation corresponding to this paper may be realized, for example, when the density of interstitials recombining with vacancies is so large that it can be regarded as a constant for local position and time or when excitons disappear due to the spontaneous radiative decay. Besides these, we hope for a future realization of experiments where the instability of excited states can be controlled spatially and temporally. The quantum nature of particles is also an attractive topic. Various extensions of our framework are expected for more realistic problems. Although the present theory does not include all actual details, we believe that these findings hold universally in phase-separation dynamics in finite-lifetime systems and will be of great significance in the interpretation of experimental results.

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*Electronic address: akira@acty.phys.sci.osaka-u.ac.jp
†Electronic address: ogawa@mailaps.org

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