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Dynamics of spinodal decomposition in finite-lifetime systems: Nonlinear statistical theory based on a coarse-grained lattice-gas model

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We study theoretically dynamics of the spinodal decomposition in *finite-lifetime* systems to clarify effects of the interparticle interactions beyond the Ginzburg-Landau-Wilson phenomenology. Our theory is based on the coarse-grained Hamiltonian derived from the interacting lattice-gas model with a finite lifetime. The information of a system is reduced to closed-form coupled integrodifferential equations for the single-point distribution function and the dynamical structure factor. These equations involve explicitly the interparticle interactions. The finite lifetime prevents the phase separation and the order formation in the cw creation case; domains cannot grow to be larger than an asymptotic characteristic size $[k_{\max}(t\rightarrow\infty)]^{-1}$. Power-law dependence of $k_{\max}(t\rightarrow\infty)$ on the interparticle interaction and the particle lifetime is also found numerically. The finite lifetime prevents the phase separation, i.e., the lower critical wave number $k_c^{(1)}$ appears and domains of size larger than $[k_c^{(1)}]^{-1}$ cannot grow.

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I. INTRODUCTION

Spinodal decomposition is a nonequilibrium phenomenon in which an unstable state separates spatially into several stable states. The spinodal decomposition has been studied since it was observed as the periodic compositional variation of metallic alloys in the 1940s (see reviews, e.g., [1–7]). It was investigated with experiments such as x-ray scattering in the 1960s and 1970s [8,9]. In the earliest theoretical works, Cahn and co-worker [10,11] and Cook [12] proposed linear theories, which describe correctly only an early stage of the phase-separation dynamics. Following them, the nonlinear theory was developed by Langer, Bar-on, and Miller (LBM) [13]. They used an approximation with which the distribution function is decoupled and the spatial correlation is described by the dynamical structure factor. One of the purposes of theoretical studies is to solve the behavior of the dynamical structure factor because it corresponds to the spectrum of x-ray, neutron, or light scatterings. These theories employed the Ginzburg-Landau-Wilson free energy. On the other hand, theories based on the kinetic spin model (i.e., Glauber's model, Kawasaki's model, etc.) have been proposed [14–16]. For example, Binder [14] derived the equation of motion for the dynamical structure factor with the use of the master-equation method from Kawasaki's spin-exchange kinetic Ising model [17]. Tomita [15] studied the mean-field dynamics based on the spherical spin model. Besides these works, many theoretical studies [18–25], computer simulations [26–29], and experiments [30–32] of the phase separation have been proposed. The phase separation is studied also in various kinds of fields such as high polymer physics [33], cosmology [34], and so on.

Thus far, almost all the theories of the spinodal decomposition are applicable only to systems with neither creation nor annihilation of constituent particles. In such systems, it is implicitly assumed that the particle has infinite lifetime. However, there are many examples where external fields create *unstable* particles (or quasiparticles) that have a *finite* lifetime and can build a new phase. Several examples of such systems are listed here: (i) a 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 [35], in this case, excited molecules are unstable; (ii) the exciton liquid phase [36] and the exciton Bose-Einstein condensation [37] created by light absorption, in which excitons have a finite radiative lifetime; (iii) the electron-hole liquid [38,39] and the electron-hole droplet [40,41] in semiconductors excited more strongly than for exciton systems, in which electron-hole pairs can disappear due to recombination within a finite lifetime, we consider the formation of these phases as the phase separation of particles with finite lifetime; (iv) new phase development and spatial modulation in crystals under nuclear and other irradiation [42–44]. Theoretical studies of the order formation such as void nucleation of vacancies were proposed [45]. They are accompanied by the creation process and the recombination process with interstitials. In these studies, however, effects of "finite lifetime" are not focused upon. 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.

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To study the phase separation in such finite-lifetime systems, we proposed theoretical formulation of the spinodal decomposition dynamics in finite-lifetime systems and discussed effects of the finite lifetime [46,47]. It was a phenomenology based on the Ginzburg-Landau-Wilson free energy and hence it had a universality for various physical systems. However, we cannot investigate effects of microscopic characteristics of the systems with its phenomenology because the microscopic information is included implicitly into the free-energy functional. Also in order to approach more realistic systems, we have to study effects of the interparticle interactions for the spinodal decomposition dynamics as well as the finite-lifetime effects. Therefore we need to construct a theory of the spinodal decomposition in finite-lifetime systems taking explicitly into account more microscopic details.

The main aim of this paper is to introduce a formalism for spinodal decomposition in finite-lifetime systems, which contains explicitly the microscopic interparticle interactions. To this end, our strategy is to start with the lattice-gas model with interparticle interactions. After a suitable coarse-graining procedure, we obtain an effective Hamiltonian of the system. The processes of the creation and the annihilation of particles are introduced in the same way as in the previous paper [46,47]. Employing an approximation to the two-point distribution function, coupled integrodifferential equations are derived in a closed form, which contains nonlinear features of the dynamics. Numerical results of these fundamental equations are examined to clarify the relation between the spinodal decomposition dynamics and the interparticle interactions in finite-lifetime systems.

This paper is organized as follows. In Sec. II, we introduce the theoretical formulation taking into account the creation and annihilation of particles starting from the coarse-grained lattice-gas model. The closed-form coupled equations of motion are derived in the LBM approximation. Our model is compared with those in other systems such as the chemically reacting systems [48–51] and the block copolymer systems [52–54]. The finite-lifetime effects on the spinodal decomposition are understood qualitatively from the Lyapunov stability analysis in Sec. III. There the critical wave numbers are introduced in order to characterize the dynamics in the early stage of the phase separation. Numerical results are shown in Sec. IV. Power-law dependence of an asymptotic characteristic domain size on the interparticle interaction and the particle lifetime is clarified numerically. Temporal development of the critical wave numbers is also discussed. Our results are compared with those in the chemically reacting systems and the block copolymer systems.

II. THEORETICAL FORMULATION

A. Coarse graining in the lattice-gas model

For the study of the spinodal decomposition, the lattice-gas model,

$$\mathcal{H}=-\tfrac{1}{2}\sum_{l_1}\sum_{l_2(\neq l_1)} V_{l_1 l_2} m_{l_1} m_{l_2}, \tag{1}$$

is often used, where l_1, l_2 are the indices of the lattice points and $V_{l_1 l_2}$ is the interaction between particles. We assume that the interparticle interaction is dependent only on the distance $|l_1-l_2|$ and is attractive, whose strength approaches zero when $|l_1-l_2|$ is large enough. Note that $|l_1-l_2|$ cannot become shorter than the lattice constant. This restriction means that the particles have hard-sphere interaction when $|l_1-l_2|$ is short enough. In this lattice-gas model, the Hamiltonian of the system is described by the particle number m_{l_1} at the lattice point l_1: $m_{l_1}=0$ or 1. To formulate the theory of the spinodal decomposition, we describe the Hamiltonian by a continuous variable, the particle density. In order to define the particle density, we have to divide the space into many small regions and count the particle number in each region. So we shall introduce the coarse-graining procedure and rewrite the lattice-gas model, Eq. (1), as the coarse-grained Hamiltonian.

We use the cell representation for the coarse-graining procedure. This procedure is that N lattice points $\{l_i=1,...,N|l_i\in i\}$ are unified to the ith cell. The system is divided into $\mathcal{N}$ pieces of the cell of size a_0. The particle number in the ith cell is written as n_i. All the lattice points $\{l_i|l_i\in i\}$ in the ith cell are assumed to have $m_{l_i}=n_i/N$ particles uniformly. With the use of this coarse graining, the lattice-gas model of Eq. (1) is transformed into the cell Hamiltonian

$$H=-\tfrac{1}{2}\sum_{i}\sum_{j(\neq i)} J_{ij} n_i n_j - K\sum_{i} n_i^2, \tag{2}$$

where

$$J_{ij}\equiv\frac{1}{N^2}\sum_{l_1\in i}\sum_{l_2\in j(\neq i)} V_{l_1 l_2} \quad (i\neq j), \tag{3}$$

$$K\equiv\frac{1}{2N^2}\sum_{l_1\in i}\sum_{l_2\in i} V_{l_1 l_2}. \tag{4}$$

This Hamiltonian is described with the intracell particle number n_i, which is $0\leqslant n_i\leqslant N$. In this paper, N is assumed to be so large that we can regard n_i as a continuous variable. Thus the intracell particle number n_i corresponds to the local density of particles. The interparticle interaction is divided into two parts: the intercell long-range interaction J_{ij} and the intracell short-range interaction K. Here J_{ij} depends only on the distance $|i-j|$. As mentioned above, the original interparticle interaction strength $|V_{l_1 l_2}|$ is small when $|l_1-l_2|\geqslant a_0$, so we consider that the strength of J_{ij} is much smaller than that of K.

B. The Fokker-Planck equation for the multipoint distribution function

We define a derivation u_i of the intracell particle number n_i from its average $n_0\equiv\langle n_i\rangle$, i.e., $u_i\equiv n_i-n_0$, as an order parameter of the system. A state of the system is described by the multipoint distribution function $P(\{u_m\},t)$, which has all statistical information of the system, where $\{u_m\}\equiv\{u_m|m=1,2,...,\mathcal{N}\}$. The spatial average of a physical quantity $O(\{u_m\})$ is evaluated as

$$\langle O(\{u_m\})\rangle(t)=\int\prod_i du_i P(\{u_m\},t)O(\{u_m\}). \tag{5}$$

Temporal evolution of $P(\{u_m\},t)$ results from two origins,

$$\frac{\partial}{\partial t}P(\{u_m\},t)=\left.\frac{\partial P(\{u_m\},t)}{\partial t}\right|_c+\left.\frac{\partial P(\{u_m\},t)}{\partial t}\right|_{\mathrm{nc}}. \tag{6}$$

The first term is the "conservation term" resulting from the particle transfer, in which the total number of particles is conserved, and the second is the "nonconservation term" coming from effects of particle creation and annihilation due to the finite lifetime.

We shall derive the first term due to the particle transfer with the use of the master-equation method by Binder [14]. The phase separation develops by the particle transfer between nearest-neighbor cells. The transition probability for this particle transfer is defined from the detailed-balance condition and depends on the energy variation. As a result, the temporal evolution of $P(\{u_m\},t)$ due to the conservation term obeys the Fokker-Planck equation (see Appendix A)

$$\begin{aligned}\left.\frac{\partial P(\{u_m\},t)}{\partial t}\right|_c=&-\frac{a_0^2}{\tau_0}\sum_i\frac{\partial}{\partial u_i}\Bigg(\Bigg[\sum_j\Delta_{ij}\Bigg\{N(n_0+u_j)\\&-\frac{J_0+2K}{k_BT}C_j(u_j,t)\Bigg\}\\&-\frac{J_0R^2}{2k_BT}\sum_j\sum_l\Delta_{ij}\Delta_{jl}C_l(u_l,t)\Bigg]P(\{u_m\},t)\\&+\sum_j\Delta_{ij}\frac{\partial}{\partial u_j}\left[\frac{\partial C_j}{\partial u_j}P(\{u_m\},t)\right]\Bigg),\end{aligned} \tag{7}$$

where

$$\begin{aligned}C_j(u_j)=&\tfrac{1}{6}n_0^2(3N-2n_0)+n_0(N-n_0)u_j+\tfrac{1}{2}(N-2n_0)u_j^2\\&-\tfrac{1}{3}u_j^3.\end{aligned} \tag{8}$$

a_0 is the volume of a cell and τ_0 is the time constant concerned with the particle transfer. k_B is the Boltzmann constant and T is the temperature. We introduce two parameters characterizing the long-range interaction between cells in the same way as Binder [14]. One is the strength J_0 defined as $J_0\equiv\Sigma_{j(\neq i)}J_{ij}$, another is the interaction range R as $R\equiv(1/\sqrt{J_0})[\Sigma_{j(\neq i)}(\mathbf{r}_j-\mathbf{r}_i)^2J_{ij}]^{1/2}$.

The creation rate of particles is denoted as $y(t)$, which is a given function of t and is assumed to be homogeneous in space, hence $y(t)$ is independent of the position $\mathbf{r}$. The particle lifetime is denoted as τ, which depends on neither t nor $\mathbf{r}$. The temporal evolution of $P(\{u_m\},t)$ due to the nonconservation term obeys the Fokker-Planck equation (see Appendix B)

$$\begin{aligned}\left.\frac{\partial P(\{u_m\},t)}{\partial t}\right|_{\mathrm{nc}}=&-\sum_i\frac{\partial}{\partial u_i}\Bigg(\left[y(t)-\frac{n_0+u_i}{\tau}\right]P(\{u_m\},t)\\&-\frac{1}{2}\frac{\partial^2}{\partial u_i^2}\left\{\left[y(t)+\frac{n_0+u_i}{\tau}\right]P(\{u_m\},t)\right\}\Bigg),\end{aligned} \tag{9}$$

which comes from the competition between particle creation and annihilation [46,47]. As a point to notice, the spatial average n_0 of the intracell particle number depends on time because of the creation and annihilation of particles, and its temporal evolutional equation is

$$\frac{d}{dt}n_0(t)=y(t)-\frac{n_0(t)}{\tau}. \tag{10}$$

Using Eqs. (6), (7), (9), and (10), we drive the full form of the Fokker-Planck equation for the multipoint distribution function $P(\{u_m\},t)$ in a system with particle creation and annihilation effects. This equation contains the interparticle interaction explicitly.

C. Closed-form coupled equations of motion: The LBM approximation

The multipoint distribution function $P(\{u_m\},t)$ has all the information of a system. But we cannot solve easily the Fokker-Planck equation for $P(\{u_m\},t)$. If it can be solved, we are afraid that essential qualities become vague. So, in our theory, the time evolution of a system is described by two quantities. One is the dynamical structure factor $S(\mathbf{k},t)$, which is defined as the Fourier transform of the two-point spatial correlation function $\langle u_\alpha u_\beta\rangle(t)$. It can be measured by x-ray, neutron, or light scattering experiments. Since we assume that the system is isotropic, $\langle u_\alpha u_\beta\rangle(t)$ depends only on the distance $|\mathbf{r}_\alpha-\mathbf{r}_\beta|$, and the definition of $S(\mathbf{k},t)$ is

$$S(\mathbf{k},t)\equiv\sum_\alpha\exp[-i\mathbf{k}\cdot(\mathbf{r}_\alpha-\mathbf{r}_\beta)]\langle u_\alpha u_\beta\rangle(t). \tag{11}$$

The other is the single-point distribution function defined as

$$P^{(1)}(u_\alpha,t)\equiv\int\prod_{i(\neq\alpha)}du_iP(\{u_m\},t). \tag{12}$$

This is the probability distribution function for the case when the order parameter in the αth cell takes the value u_α at time t. Since α is arbitrary ($\alpha=1,2,\ldots,\mathcal{N}$), $P^{(1)}(u_\alpha,t)$ also denotes the distribution function of the number of cells in which the particle number is $n_\alpha=u_\alpha+n_0$. Using only $P^{(1)}(u_\alpha,t)$, the average of a quantity $O(u_\alpha)$, which is a function only of u_α (not of u_β), is calculated as

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

In order to derive closed-form coupled equations for the single-point distribution function $P^{(1)}(u,t)$ and the dynamical structure factor $S(\mathbf{k},t)$ from Eqs. (6), (7), and (9), we employ an approximation [13], where the two-point distribution function is decoupled as

$$P^{(2)}(u_\alpha,u_\beta,t)=P^{(1)}(u_\alpha,t)P^{(1)}(u_\beta,t)\times\left[1+\frac{\langle u_\alpha u_\beta\rangle(t)}{[\langle u^2\rangle(t)]^2}u_\alpha u_\beta\right], \tag{14}$$

where α and β stand for two different cells. In this paper, we call this approximation the LBM approximation. Moreover we introduce the following terms. N for the particle number, a_0 for length, τ_0 for time, the critical temperature $k_BT_c\equiv(N/4)(J_0+2K)$, and k_BT_c/N as the interaction coefficient. After this normalization, we should note that the relation $J_0+2K=4$ always holds. Applying the LBM approximation and the above-mentioned normalization, the closed-form coupled integrodifferential equations for $P^{(1)}(u,t)$ and $S(\mathbf{k},t)$ are obtained as

$$\begin{aligned}\frac{\partial}{\partial t}P^{(1)}(u,t)=\frac{\partial}{\partial u}\Bigg[\Bigg(&N\Bigg\{D_f^*(T)[C-\langle C\rangle(t)-B(t)u]\\&+\frac{X(t)}{\langle u^2\rangle(t)}u\Bigg\}-\left[y(t)-\frac{n_0+u}{\tau}\right]\Bigg)P^{(1)}(u,t)\\&+D_f^1\frac{\partial}{\partial u}\left[\frac{\partial C}{\partial u}P^{(1)}(u,t)\right]\\&+\frac{1}{2N}\frac{\partial}{\partial u}\left\{\left[y(t)+\frac{n_0+u}{\tau}\right]P^{(1)}(u,t)\right\}\Bigg],\end{aligned} \tag{15}$$

$$\begin{aligned}\frac{\partial}{\partial t}S(\mathbf{k},t)=&-2\left\{Nk^2\left[1-\frac{4}{T}B(t)+\frac{J_0R^2}{2T}B(t)k^2\right]+\frac{1}{\tau}\right\}S(\mathbf{k},t)\\&+\frac{2}{v_0}[n_0-n_0^2-\langle u^2\rangle(t)]k^2+\frac{1}{Nv_0}\left[y(t)+\frac{n_0}{\tau}\right],\end{aligned} \tag{16}$$

where N is an upper limit of the particle number in one cell and $v_0\equiv a_0^3$ is the volume of a cell. The functions C, $B(t)$, $X(t)$, and $D_f^*(T)$ are defined as

$$\begin{aligned}C=C(u)\equiv&\tfrac{1}{6}n_0^2(3-2n_0)+n_0(1-n_0)u\\&+\tfrac{1}{2}(1-2n_0)u^2-\tfrac{1}{3}u^3,\end{aligned} \tag{17}$$

$$B(t)\equiv n_0(1-n_0)+\frac{1}{2}(1-2n_0)\frac{\langle u^3\rangle(t)}{\langle u^2\rangle(t)}-\frac{1}{3}\frac{\langle u^4\rangle(t)}{\langle u^2\rangle(t)}, \tag{18}$$

$$X(t)\equiv\frac{1}{(2\pi)^3}\int d\mathbf{k}\,k^2\left[1-\frac{4}{T}B(t)+\frac{J_0R^2}{2T}B(t)k^2\right]S(\mathbf{k},t), \tag{19}$$

$$D_f^*(T)\equiv-D_f^1\frac{4}{T}+D_f^2\frac{J_0R^2}{2T}. \tag{20}$$

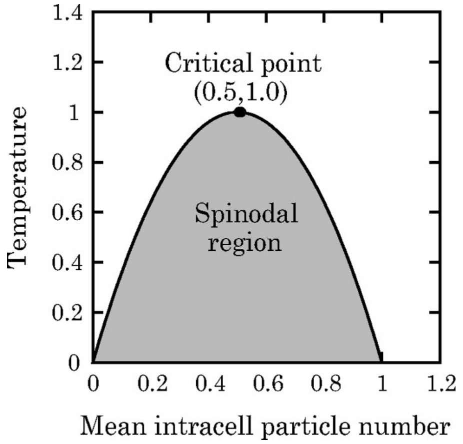

FIG. 1. The spinodal line drawn on the $(n_0/N,T/T_c)$ plane. The critical temperature and the critical intracell particle number are given as $T_c=1.0$ and $n_c/N=0.5$, respectively. The spinodal region is shadowed. We use the following terms: N for mean intracell particle number and T_c for temperature.

Here the $\mathbf{k}$ integration is carried out in the range of $0\leqslant|\mathbf{k}|\leqslant v_0^{-1/3}$, and $-D_f^1=-(2\pi)^{-3}\int d\mathbf{k}\,k^2$ and $D_f^2=(2\pi)^{-3}\times\int d\mathbf{k}\,k^4$ are the diagonal elements of the normalized Laplacian operators. In these equations, the interparticle interaction appears as J_0R^2. As mentioned above, the short-range part K is connected with the long-range one J_0 by the relation $K=2-\frac{1}{2}J_0$. Recall that the mean intracell particle number n_0 depends on time according to Eq. (10). The temporal development of the spinodal decomposition in many-particle systems with creation and annihilation is described with these equations. But there are conditions for the occurrence of the spinodal decomposition. The spinodal line is given as $T=4n_0(1-n_0)$, which is shown in Fig. 1. The spinodal decomposition can occur only when the initial state is within the shadowed region

$$T<1 \quad\text{and}\quad \tfrac{1}{2}(1-\sqrt{1-T})<n_0<\tfrac{1}{2}(1+\sqrt{1-T}). \tag{21}$$

D. Comparison of our model with other systems

The temporal evolution of the order parameter $u(\mathbf{r},t)$ due to the nonconservation term obeys the equation

$$\left.\frac{\partial}{\partial t}u(\mathbf{r},t)\right|_{\mathrm{nc}}=y(t)-\frac{n_0+u(\mathbf{r},t)}{\tau}.$$

This describes the competition between the creation $[y(t)]$ and the annihilation $[\tau]$. Here we shall discuss similarity and difference between our model and others, e.g., chemically reacting systems [48–51] and block copolymer systems [52–54]. The time-evolutional equations for an order parameter in these systems are equivalent *mathematically* to those in our model. However *physical* origins are quite different from each other. In chemically reacting systems, the components are created and annihilated by chemical reactions. The time-evolutional equation of the order parameters has the linear or nonlinear source term indicating the chemical reaction. In the case of a $A \overset{\eta}{\rightleftharpoons} B$ type reaction, for example, the equation of the order parameter $\phi(\mathbf{r},t)$, the density of A (or B) molecules has the linear source term

$$\frac{\partial}{\partial t}\phi(\mathbf{r},t)\bigg|_{\text{linear source}} = -\eta\phi + \eta(1-\phi),$$

where η is the chemical reaction rate. In the block copolymer systems, the free-energy functional has two terms, the usual Ginzburg-Landau-Wilson free-energy functional and the long-range repulsive interaction part. As a result of the long-range repulsive interaction part, the equation of the order parameter $\phi(\mathbf{r},t)$, the density of polymers, has the subtractive term

$$\frac{\partial}{\partial t}\phi(\mathbf{r},t)\bigg|_{\text{repulsion}} = -G\phi(\mathbf{r},t),$$

where G is the parameter concerned with the strength of the long-range repulsive interaction. The parameters η and G correspond to τ^{-1} in our model.

III. LYAPUNOV STABILITY ANALYSIS

Before solving Eqs. (15) and (16) numerically, we shall discuss qualitatively the effects of finite lifetime. The Lyapunov spectrum has often been used in order to discuss the stability of the system. When we assume that the fluctuation mode $u_{\mathbf{k}}(t)$ with wave number $\mathbf{k}$ of the order parameter is written as

$$u_{\mathbf{k}}(t) \propto \exp[t\Gamma_{\mathbf{k}}(t)],$$

$\Gamma_{\mathbf{k}}(t)$ is called the Lyapunov spectrum. It means the growing speed of a fluctuation mode $u_{\mathbf{k}}$ with wave number $\mathbf{k}$ in the early state. The fluctuation mode grows if $\Gamma_{\mathbf{k}}(t) > 0$ and decays if $\Gamma_{\mathbf{k}}(t) < 0$. The Lyapunov spectrum describes also the increase or decrease of the dynamical structure factor through the relation $S(\mathbf{k},t) = |u_{\mathbf{k}}(t)|^2 \propto \exp[2t\Gamma_k(t)]$ when the system is isotropic and translational symmetric. The wave number $\mathbf{k}$ is replaced simply by its amplitude $k \equiv |\mathbf{k}|$.

In the case of $y(t) = n_0/\tau = \text{const}$, the Lyapunov spectrum of the early state is derived as

$$\Gamma_k(t) = -\frac{NJ_0R^2}{2T}B(t)k^2[k^2 + A(t)] - \frac{1}{\tau}, \tag{22}$$

where

$$A(t) \equiv \frac{2}{J_0R^2}\left[\frac{T}{B(t)} - 4\right] \tag{23}$$

from Eq. (16). The forms of the Lyapunov spectra at time t are shown in Fig. 2 as a function of the wave number k. In these figures, we can define the critical wave numbers as a boundary wave number between unstable $[\Gamma_k(t) > 0]$ and stable $[\Gamma_k(t) < 0]$ fluctuation modes.

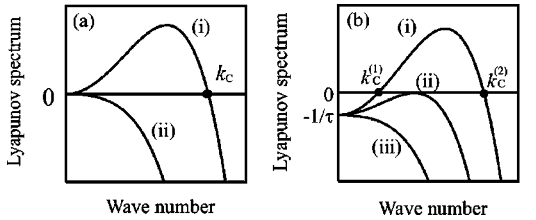

FIG. 2. Schematic view of the Lyapunov spectrum $\Gamma_{\mathbf{k}}$ as a function of $|\mathbf{k}|$. The case of neither creation $[y(t)=0]$ nor annihilation $[\tau \to \infty]$ is shown in (a), and the case of cw creation and annihilation $[y(t) = n_0/\tau = \text{const}]$ is given in (b). In (a), two lines show the cases where (i) $A(t) < 0$ and (ii) $A(t) \geqslant 0$. In (b), three lines show the cases where (i) $A(t) < A_c$, (ii) $A(t) = A_c$, and (iii) $A(t) > A_c$. Here $A_c \equiv (2/N\tau)[1 - (1 + 8N\tau/J_0R^2)^{1/2}]$. A unit for the wave number is a_0^{-1}.

In the case with neither creation nor annihilation of particles $[y(t) = 0$ and $\tau \to \infty]$, only *one* critical wave number $k_c(t)$ can be defined as

$$k_c(t) \equiv \sqrt{-A(t)} \tag{24}$$

from Fig. 2(a). The fluctuation modes with the larger wavelength than $[k_c(t)]^{-1}$ grow but those with the smaller wavelength decay. This corresponds with the usual spinodal decomposition. In the case of $y(t) = n_0/\tau = \text{const}$, we can find *two* critical wave numbers from Fig. 2(b). One is the lower critical wave number $k_c^{(1)}(t)$ and the other is the upper critical wave number $k_c^{(2)}(t)$, whose values are

$$k_c^{(1)}(t) = \frac{1}{\sqrt{2}}\left(-A(t) - \left[[A(t)]^2 - \frac{4}{N_T}A(t) - \frac{32}{NJ_0R^2\tau}\right]^{1/2}\right)^{1/2} \tag{25}$$

$$k_c^{(2)}(t) = \frac{1}{\sqrt{2}}\left(-A(t) + \left[[A(t)]^2 - \frac{4}{N\tau}A(t) - \frac{32}{NJ_0R^2\tau}\right]^{1/2}\right)^{1/2}. \tag{26}$$

Only the fluctuation modes with the wave number $k_c^{(1)}(t) < k < k_c^{(2)}(t)$ can grow, and the modes with the smaller wave number $k < k_c^{(1)}(t)$ decay in time. This appearance of decaying fluctuation modes with a smaller wave number $k < k_c^{(1)}(t)$ is one of the effects of particle creation and annihilation. If $y(t) = 0$ and $\tau \to \infty$, $k_c^{(1)}(t)$ becomes zero and $k_c^{(2)}(t)$ becomes $k_c(t) = \sqrt{-A(t)}$.

IV. NUMERICAL RESULTS AND DISCUSSION

Numerical calculation for Eqs. (15) and (16) is carried out *without* the use of the double-Gaussian ansatz in [13]. In this section, numerical results are explained and effects of the interparticle interaction and finite lifetime are discussed. In this paper, we confine ourselves to 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,

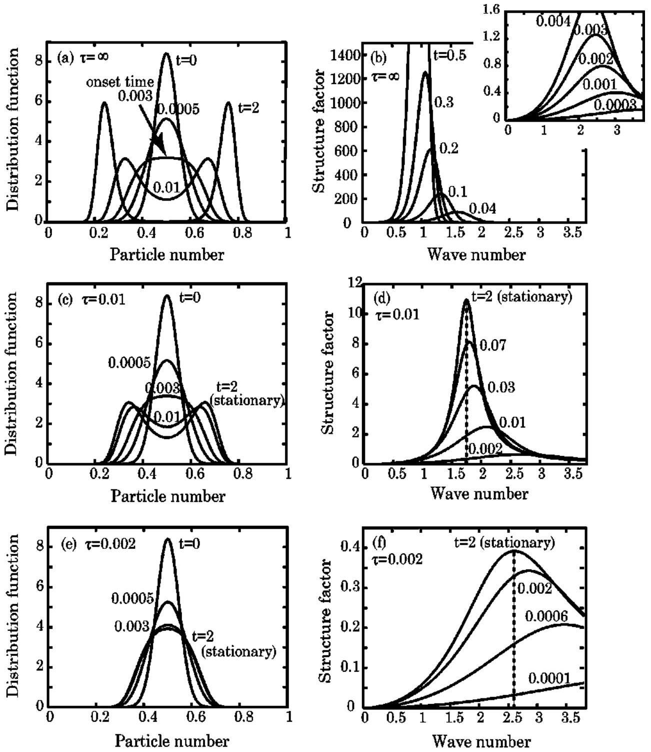

FIG. 3. Temporal development of the single-point distribution function $P^{(1)}(u,t)$ and the dynamical structure factor $S(\mathbf{k},t)$ for $T=0.9$, $N=1000$, $v_0=0.01$, and $n_0=0.5$. The creation rate is $y=n_0/\tau=0.5/\tau$ and the long-range part of interaction is $J_0R^2=0.08$. Three figures in the left row, (a), (c), and (e), are $P^{(1)}(u,t)$ and ones in the right row, (b), (d), and (f), are $S(\mathbf{k},t)$. The lifetime of particles is $\tau=\infty$ for (a) and (b), 0.01 for (c) and (d), and 0.002 for (e) and (f). The inset of (b) shows the behavior during $0\leqslant t\lesssim 0.004$. In (c)–(f) for the case of the finite lifetime, the states with "$t=2$ (stationary)" can be considered as stationary ones. Terms are defined as following: N for particle number and a_0^{-1} for wave number.

that is, both y and the mean intracell particle number $n_0=y\tau$ are constant in time. We set the parameters $N=1000$ and $v_0=0.01$. They are concerned with the coarse-graining procedures, so N and v_0 should not be so small and so large, respectively. If these conditions for N and v_0 are satisfied, final numerical results are insensitive to values of N and v_0. We set temperature $T=0.9$ and the mean intracell particle number $n_0=0.5$, which exist in the spinodal region. The initial state at $t=0$ is chosen to be a uniform-density state. Then the initial single-point distribution function $P^{(1)}(u,t=0)$ has a delta-function-like form with a small width due to the thermal fluctuation. In numerical calculations, we assume it to be Gaussian, whose variance is 0.002 25. The boundary conditions for the single-point distribution function are $P^{(1)}(u,t)|_{u<-n_0}=0$ and $P^{(1)}(u,t)|_{u>N-n_0}=0$, which result from the fact that the intracell particle number should be positive and that it must be smaller than N. The initial dynamical structure factor is assumed to be $S(\mathbf{k},t=0)=0$, which reflects that no domain structure exists at $t=0$. Since the total particle number is conserved, the boundary condition for the dynamical structure factor is $S(\mathbf{k},t)|_{\mathbf{k}=0}=0$. The largest normalized wave number should be $(6\pi^2)^{1/3}$, which comes from the fact that the number of states is equal to the number of cells.

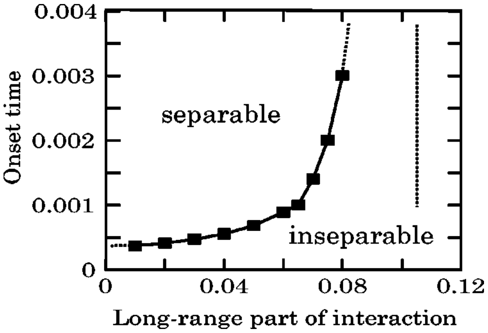

FIG. 4. Relation between the onset time and the long-range part of interaction J_0R^2. When J_0R^2 is fixed, the phase separation and the order formation cannot occur if the particle lifetime is less than the onset time, in which case it is described as "inseparable." We use the following terms: $k_BT_ca_0^2/N$ for the long-range part of interaction J_0R^2 and τ_0 for the onset time.

A. Single-point distribution function and dynamical structure factor

When the long-range part of the interaction is $J_0R^2=0.08$, temporal evolutions of the single-point distribution function and the dynamical structure factor are shown in Fig. 3. First, we shall discuss the case of $\tau=\infty$ (correspondingly $y=0$), as shown in Figs. 3(a) and 3(b). From these figures, we find that the initial state with one uniform density separates into two states with different densities. Here we define the "onset time" t_{onset} as the time nearly when one peak of $P^{(1)}(u,t)$ separates into two peaks. In Fig. 3(a), when the long-range part of the interaction is $J_0R^2=0.08$, the onset time is $t_{\mathrm{onset}}\simeq 0.003$. According to numerical calculations for different values of J_0R^2, we obtain a relation between t_{onset} and J_0R^2, as shown in Fig. 4. We find that smaller the J_0R^2 is larger is the short-range part $K=2-\frac{1}{2}J_0$ and shorter the t_{onset} becomes rapidly. This relation comes from the fact that many small domains are rapidly formed if the attractive short-range interaction is large. Moreover, there seems to exist an asymptotic upper limit of J_0R^2 for occurrence of the phase separation. Temporal evolution of the dynamical structure factor is shown in Fig. 3(b). From this figure, we find that the characteristic wave number $k_{\max}(t)$, at which the dynamical structure factor takes the maximum value, shifts to the longer-wavelength side. When $t\to\infty$, the form of $S(\mathbf{k},t\to\infty)$ may become the Lorentzian form with a maximum peak at $\mathbf{k}=0$, i.e., $k_{\max}(t\to\infty)=0$, where the system separates completely into two phases.

Next, we discuss the case of finite lifetime, as shown in Figs. 3(c)–3(f). Temporal evolutions of $P^{(1)}(u,t)$ are shown in (c) and (e). From Figs. 3(c) and 3(e), it is found that the phase separation or the order formation takes place if the lifetime ($\tau=0.01$) is larger than the onset time ($t_{\mathrm{onset}}\simeq 0.003$). In fact, when $\tau=0.002$ is shorter than the onset time, the phase separation cannot occur as shown in Fig. 3(c). Thus an effect of the finite lifetime is to prevent the phase separation or the order formation. We can interpret that

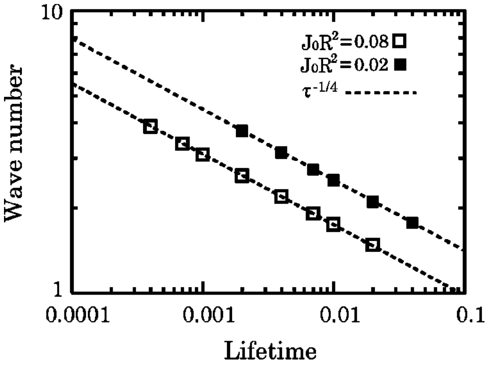

FIG. 5. The power-law relation between the particle lifetime τ and the asymptotic (stationary) value $k_{\mathrm{max}}(t \to \infty)$ of the characteristic wave number of $S(\mathbf{k},t)$. The solid points correspond to the case of $J_0R^2 = 0.02$ and the open points to $J_0R^2 = 0.08$. Both broken lines are proportional to $\tau^{-1/4}$. The term for the lifetime is τ_0 and one for wave number is a_0^{-1}.

Fig. 4 is a diagram showing a critical lifetime between being "separable" and "inseparable" into two phases for various interparticle interactions J_0R^2. Temporal evolutions of $S(\mathbf{k},t)$ are, on the other hand, shown in Figs. 3(d) and 3(f). The growth of $S(\mathbf{k},t)$ stops in the case of the finite lifetime. The stationary form of $S(\mathbf{k},t)$ is broad when the lifetime is short enough. This indicates that domains cannot grow to a sufficiently large size and that the random thermal-fluctuation modes are dominant. In addition, $k_{\mathrm{max}}(t)$ settles into a finite wave number $k_{\mathrm{max}}(t \to \infty)$ as $t \to \infty$. This is interpreted as follows: the system cannot separate completely and that domains of the finite size $[k_{\mathrm{max}}(t \to \infty)]^{-1}$ finally remain.

We find numerically two power laws for $k_{\mathrm{max}}(t \to \infty)$. One is between $k_{\mathrm{max}}(t \to \infty)$ and the particle lifetime τ as

$$k_{\mathrm{max}}(t \to \infty) \propto \tau^{-1/4}, \tag{27}$$

which is clearly shown in Fig. 5. The power $-\frac{1}{4}$ is the same as that in our previous theory [47] and is independent of the interaction J_0R^2. The other is the relation between $k_{\mathrm{max}}(t \to \infty)$ and the long-range part of the interaction J_0R^2 as

$$k_{\mathrm{max}}(t \to \infty) \propto (J_0R^2)^{-1/4}, \tag{28}$$

which is confirmed by Fig. 6. The power $-\frac{1}{4}$ is independent of the particle lifetime τ. These power laws, Eqs. (27) and (28), reflect the fact that particles within the wider range cannot converge before annihilation if the particle lifetime is short enough or if the long-range part of the interparticle interaction is weak enough.

B. The critical wave numbers

Figure 7 shows the temporal development of the critical wave numbers for the cases (a) $\tau = \infty$ and (b) $\tau = 0.01$. In the case (a) of $\tau = \infty$, there is only one critical wave number $k_c(t)$. The shadowed region denotes the unstable fluctuation modes. In other words, the fluctuation modes with the smaller wave number $k < k_c$ grow, but those with the larger wave number $k > k_c$ decay. This is the mechanism of the well-known spinodal decomposition.

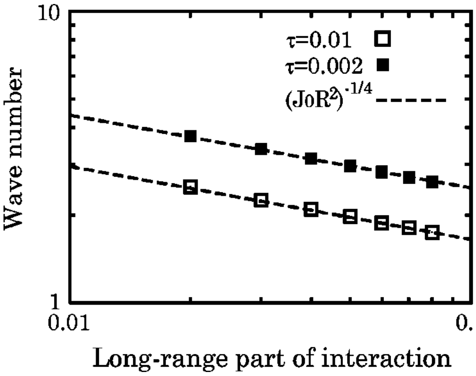

FIG. 6. The power-law relation between the long-range part of interaction J_0R^2 and the asymptotic (stationary) value $k_{\mathrm{max}}(t \to \infty)$ of the characteristic wave number of $S(\mathbf{k},t)$. The solid points correspond to the case of $\tau = 0.002$ and the open points to $\tau = 0.01$. Both broken lines are proportional to $(J_0R^2)^{-1/4}$. The terms used are defined as following: $k_BT_ca_0^2/N$ for the long-range part of interaction J_0R^2 and a_0^{-1} for wave number.

In the case (b) of $\tau = 0.01$, there are two critical wave numbers: the lower critical wave number $k_c^{(1)}(t)$ and the upper critical wave number $k_c^{(2)}(t)$. The shadowed region shows the unstable fluctuation modes. In short, the fluctuation modes only with the wave number $k_c^{(1)}(t) < k < k_c^{(2)}(t)$ can grow and other modes decay. The main difference from the case (a) of $\tau = \infty$ is the appearance of the lower critical wave number $k_c^{(1)}(t)$ and of the decaying modes with the smaller wave number $k < k_c^{(1)}$.

C. Comparison of our model with other systems

As mentioned in Sec. II D, there are similarities between our systems and other systems such as the chemically reacting systems and the block copolymer systems. The chemical reaction rate η and the strength of long-range repulsive interaction between copolymers G correspond to the lifetime of particles τ^{-1} in our model and they prevent the phase separation. This restraint of the phase separation in these systems is often called "pinning or freezing" of the phase separation. The power laws $L \propto (1/\eta)^{\alpha_1}$ and $L \propto G^{\alpha_2}$ have

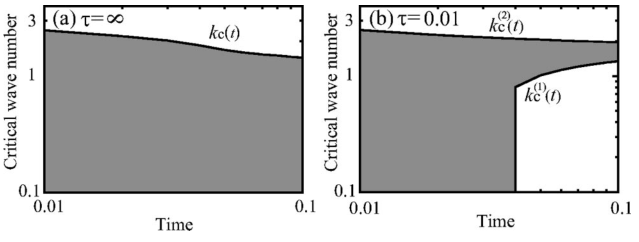

FIG. 7. Temporal development of the critical wave numbers for (a) $\tau = \infty$ and (b) $\tau = 0.01$. In (a), there is only one critical wave number $k_c(t)$. In (b), on the other hand, there are two critical wave numbers, $k_c^{(1)}(t)$ and $k_c^{(2)}(t)$. Only the fluctuation modes within the shadowed region are unstable and can grow. The appearance of the stable modes of smaller wave number $k < k_c^{(1)}(t)$ is found in (b). We use the following terms: τ_0 for time and a_0^{-1} for the critical wave number.

been also derived numerically [48,52,53], where L is the asymptotic domain or pattern size in the stationary state when time $t \rightarrow \infty$.

Moreover, in these systems, the lower critical wave number also appears, and the modes with wave numbers smaller than the lower critical wave number decay [48–50]. The appearance of the lower critical wave number and the restraint of the fluctuation growth with the longer wavelength is a result common to phase separating systems far from equilibrium [55].

V. DISCUSSION AND CONCLUSIONS

We have formulated a theoretical model of the spinodal decomposition dynamics in the finite-lifetime system. Our model includes effects of the interparticle interaction explicitly, in contrast to the Ginzburg-Landau-Wilson phenomenology. The nonlinear coupled equations for $P^{(1)}(u,t)$ and $S(\mathbf{k},t)$ have been derived with the master-equation method and the LBM approximation. In general, effects of finite lifetime prevent occurrence of the phase separation and the order formation. We have discussed the onset time, i.e., the boundary lifetime, which decides whether or not the phase separation occurs. Domains with a finite size $[k_{\max}(t \rightarrow \infty)]^{-1}$ are found to remain finally, whose size is proportional to $\tau^{1/4}$ and $(J_0 R^2)^{1/4}$. We discussed the temporal development of two critical wave numbers. One of these critical wave numbers, the lower critical wave number $k_c^{(1)}(t)$, appears as a result of finite lifetime. Similar characteristics are shown in chemically reacting systems and block copolymer systems.

As a final discussion in this paper, we make comments on the unsolved problems of our theory. In our formulation, the phase separation can occur only when the longer-wavelength modes increase, i.e., the state that corresponds to that within the spinodal region. However, for practical purposes, the phase separation can occur even out of the spinodal region via the nucleation mechanisms. In order to describe the nucleation, the volume and surface effects of domains should be adopted into the particle-transfer probability, and we have to obtain more detailed spatial infirmations. As another problem, we should clarify what is the origin of the power laws for the asymptotic domain size $[k_{\max}(t \rightarrow \infty)]^{-1}$. To this end, we need to solve the equations for $P^{(1)}(u,t)$ and $S(\mathbf{k},t)$ analytically, or derive the power directly with other microscopic models. The quantum nature of particles is also an attracting topic. We are interested in the more detailed and microscopic mechanism of phase separation and order formation. In this paper, we have given one example of the universality of finite-lifetime effects on the spinodal decomposition.

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APPENDIX A: THE TEMPORAL EVOLUTION OF THE MULTIPOINT DISTRIBUTION FUNCTION DUE TO THE CONSERVATION TERM

The conservation term due to the particle transfer is derived with the use of the master-equation method by Binder [14]. The phase separation progresses by the particle transfer between nearest-neighbor cells. For example, we consider the particle transfer from the ith cell to the α_ith cell and then the configuration of particles changes from $\{n_1,\ldots,n_i,n_{\alpha_i},\ldots,n_{\mathcal{N}}\}$ to $\{n_1,\ldots,n_i-1,n_{\alpha_i}+1,\ldots,n_{\mathcal{N}}\}$. When the transition probability for this configuration change is denoted as $W(n_i \rightarrow n_i-1, n_{\alpha_i} \rightarrow n_{\alpha_i}+1)$, the temporal evolution of $P(\{n_m\},t)$ obeys the master equation

$$\begin{aligned}\left.\frac{\partial P(\{n_m\},t)}{\partial t}\right|_c = & \sum_i \sum_{\alpha_i} W(n_i-1 \rightarrow n_i, n_{\alpha_i}+1 \rightarrow n_{\alpha_i}) \\ & \times P(n_1,\ldots,n_i-1,n_{\alpha_i}+1,\ldots,n_{\mathcal{N}},t) \\ & - \sum_i \sum_{\alpha_i} W(n_i \rightarrow n_i-1, n_{\alpha_i} \rightarrow n_{\alpha_i}+1) \\ & \times P(\{n_m\},t). \end{aligned} \tag{A1}$$

The summation Σ_i runs over all $\mathcal{N}$ pieces of cells and Σ_{α_i} runs over the nearest-neighbor cells of the ith cell. Here $P(n_1,\ldots,n_i-1,n_{\alpha_i}+1,\ldots,n_{\mathcal{N}},t)$ is the probability for the realization of a particle configuration $\{n_1,\ldots,n_i-1,n_{\alpha_i}+1,\ldots,n_{\mathcal{N}}\}$ at time t. The transition probability should meet the detailed-balance condition

$$\begin{aligned} W(n_i-1 \rightarrow n_i, n_{\alpha_i}+1 \rightarrow n_{\alpha_i}) P_{\mathrm{eq}}(n_1,\ldots,n_i-1,n_{\alpha_i}+1,\ldots,n_{\mathcal{N}}) \\ = W(n_i \rightarrow n_i-1, n_{\alpha_i} \rightarrow n_{\alpha_i}+1) P_{\mathrm{eq}}(\{n_m\}). \end{aligned}$$

$P_{\mathrm{eq}}(\{n_m\})$ is the multipoint distribution function in a thermal equilibrium state and it is defined as

$$P_{\mathrm{eq}}(\{n_m\}) \equiv Z^{-1} Y(\{n_m\}) \exp\left[-\frac{H(\{n_m\})}{k_B T}\right]. \tag{A2}$$

Here Z is the partition function, $H(\{n_m\})$ is the coarse-grained Hamiltonian, Eq. (2), for the particle configuration $\{n_m\}$, and $Y(\{n_m\})$ comes from the information of the original lattice; $Y(\{n_m\}) = {}_N C_{n_1} \cdots {}_N C_{n_i}\, {}_N C_{n_{\alpha_i}} \cdots {}_N C_{n_{\mathcal{N}}}$. This factor, $Y(\{n_m\})$, is related to the number of the original microscopic states with energy $H(\{n_m\})$. Using this equilibrium distribution function, one of the transition probabilities satisfying the detailed-balance condition is defined as

$$W(n_i \to n_i - 1, n_{\alpha_i} \to n_{\alpha_i} + 1) = \frac{1}{2\tau_0} n_i (N - n_{\alpha_i}) \times \left\{ 1 - \tanh\left[\frac{\delta H(n_i \to n_i - 1, n_{\alpha_i} \to n_{\alpha_i} + 1)}{2k_B T} \right] \right\}. \tag{A3}$$

$\delta H(n_i \to n_i - 1, n_{\alpha_i} \to n_{\alpha_i} + 1)$ is the variation of energy from the particle transfer and τ_0 is the shortest time for a particle transfer between nearest-neighbor cells. The factor $n_i(N - n_{\alpha_i})$ in the transition probability comes from the fact that one cell has N pieces of the original lattice points. Thus the master equation, Eqs. (A1) and (A3), for $P(\{n_m\},t)$ is obtained.

This master equation is rewritten as the Fokker-Planck equation for the multipoint distribution function $P(\{u_m\},t)$. For this procedure, the energy variation δH is assumed to be much smaller than the temperature $k_B T$, then $\tanh(\delta H/2k_B T) \approx \delta H/2k_B T$. Next, the functions of $n_m \pm 1$ are expanded up to the second order,

$$f(n_m \pm 1) \approx f(n_m) \pm \frac{\partial}{\partial n_m} f(n_m) + \frac{1}{2} \frac{\partial^2}{\partial n_m^2} f(n_m), \tag{A4}$$

and the space functions are also expanded to the second order of a_0

$$f(n_{\alpha_i}) \approx f(n_i) + a_0^2 \sum_j \Delta_{ij} f(n_j). \tag{A5}$$

Two parameters characterizing the long-range interaction, Eq. (3), are introduced in the same way as Binder [14]; One is the strength J_0 and another is the interaction range R. Finally, we rewrite the variable n_m as the order parameter $u_m \equiv n_m - n_0$. Applying these procedures to the master equation, Eqs. (A1) and (A3), we can derive the Fokker-Planck equation for $P(\{u_m\},t)$, Eq. (7).

APPENDIX B: THE TEMPORAL EVOLUTION OF THE MULTIPOINT DISTRIBUTION FUNCTION DUE TO THE NONCONSERVATION TERM

The temporal evolution of $P(\{n_m\},t)$ due to the nonconservation term obeys the master equation

$$\left.\frac{\partial P(\{n_m\},t)}{\partial t}\right|_{\mathrm{nc}} = \sum_i \left[y(t) P(n_1,\ldots,n_i - 1,\ldots,n_{\mathcal{N}},t) - y(t) P(\{n_m\},t) + \frac{1}{\tau}(n_i + 1) P(n_1,\ldots,n_i + 1,\ldots,n_{\mathcal{N}},t) - \frac{1}{\tau} n_i P(\{n_m\},t) \right], \tag{B1}$$

which comes from the competition between particle creation and annihilation [46,47]. Here $P(n_1,\ldots,n_i \pm 1,\ldots,n_{\mathcal{N}},t)$ is the probability distribution function for the particle configuration $\{n_1,\ldots,n_i \pm 1,\ldots,n_{\mathcal{N}}\}$. The summation Σ_i runs over all $\mathcal{N}$ pieces of cells. Since n_m is assumed to be a continuous variable, the function of $n_m \pm 1$ is expanded to the second order as in Eq. (A4). Rewriting the variable n_m as the order parameter u_m, we derive the Fokker-Planck equation for the multipoint distribution function $P(\{u_m\},t)$ due to particle creation and annihilation, Eq. (9).

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