

## **Aspects of Nucleation and Drift Processes in a One-Dimensional Model**

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We introduce a one-dimensional model involving the nucleation and the drift of many particles. The model originates from interacting kink systems and simulates time evolution in modulated systems. In this model the nucleation rate of a particle depends nonlocally on the density of preexisting particles and the drift of particles is due to a weak and repulsive interaction among them. We first study the statistics of this model in the case that the drift of particles is negligible, and then consider the effects of the drift of particles.

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**KEY WORDS:** Statistical dynamics; transient process; nucleation; drift; one dimension.

### **1. INTRODUCTION**

We often observe nucleation and/or drift of some topological defects in a system during transition from one stationary or equilibrium state to another. For example, the commensurate to incommensurate (C-IC) phase transition<sup>(1)</sup> requires the nucleation and growth of so-called stripples, i.e., particular clusters of discommensurations. Also, the smectic C to smectic C\* (SmC-SmC\*) transition in a thin liquid crystal layer,<sup>(2)</sup> which is induced by removing an external electric field, involves the appearance of straight, twisted disclinations and their motion. Other examples occur in the convection of fluids and in the plastic deformation of solids, and so on.

In this paper we study a simplified one-dimensional model involving nucleation and/or drift processes of particles (by which we mean topological defects) in one dimension. In our model we take into account the following two properties, which are often shared by real systems: (1) The rate (nucleation rate) at which a new particle appears per unit time

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and per unit length of the system depends on the local density of already existing particles.<sup>(2)</sup> (2) The interaction between the particles is repulsive and of very short range compared with the distance between particles, and the motion of a particle due to the interaction is overdamped by friction with the background. In Section 2 we define the model, which involves both the nucleation and the drift of particles.<sup>(3)</sup> This model is a microscopic model in which each particle preserves its identity.

In order to derive the hydrodynamic theory for the density fluctuation of particles from this model one has to assume that the particle distribution is locally stationary in a short time interval. The validity of this assumption, however, is severely restricted in our model when the drift of particles is taken into account. Hence we first study in Section 3 the hydrodynamic theory of the model without the interaction between particles. Detailed calculation is done for a concrete example (Appendix D). The correlation hole is found in the (macroscopic) density correlation function, which reflects the saturation effect of the nucleation rate, and the size of the correlation hole is shown to be time-dependent.

Then in Section 4 we study drift of the particles with no nucleation but from two initial configurations that typify the transient distributions of the particles in the nucleation-plus-drift system. When the interaction between particles is strongly nonlinear, we find (1) that a cluster of particles diffuses by two modes, first by smearing out of the boundaries of the cluster and then by spreading out of the cluster itself with much a slower time dependence than the former, and (2) that a transient shock front is formed when a particle is added to the almost periodic array of the particles. We also argue that a hydrodynamic theory for the nucleation-plus-drift process is unlikely to be established.

Section 5 is devoted to a summary.

## 2. MICROSCOPIC MODEL

We denote the position of the  $n$ th particle on the  $x$  axis by  $x_n(t)$  such that

$$\dots < x_n(t) < x_{n+1}(t) < \dots \quad (2.1)$$

where  $t$  is the time. The interaction energy between the two particles at a distant  $\Delta$  is denoted by  $V(\Delta/\xi)$ , where  $\xi$  is the range of the interaction and  $V(z)$  is a decreasing function of  $z$  since we assume that the interaction is repulsive. In real systems the range of interaction  $\xi$  is of the order of the particle size, i.e., the typical width of a topological defect such as a discommensuration or a twisted disclination. In the following we study only the

case where the interparticle distances are always much larger than  $\xi$  (otherwise the particles lose their identity), and the interaction  $V(A/\xi)$  is strongly nonlinear for  $A/\xi \gg 1$ . Neglecting the thermal fluctuation force other than that is responsible for the nucleation, we describe the overdamped drift motion of particles as follows:

$$\frac{d}{dt} x_n = -\Gamma \frac{\partial}{\partial x_n} \left[ \frac{1}{2} \sum_l \sum_m V \left( \frac{|x_l - x_m|}{\xi} \right) \right] \quad (2.2)$$

$$\simeq -\frac{\Gamma}{\xi} \left[ V' \left( \frac{x_n - x_{n-1}}{\xi} \right) - V' \left( \frac{x_{n+1} - x_n}{\xi} \right) \right] \quad (2.3)$$

where  $\Gamma$  is inverse of the friction constant for the particle motion and  $V'(z) = dV(z)/dz$ . The form of (2.2) or (2.3) is unsuitable for describing the nucleation, since upon nucleation the number of variables changes and a renumbering of their indices is required. By introducing the field variable we can avoid such trouble. Defining the microscopic density of particles by

$$\hat{\rho}(x, t) = \sum_n \delta(x - x_n(t)) \quad (2.4)$$

we can rewrite (2.2) in the following form (this model was introduced in further detail in Ref. 3):

$$\left. \frac{\partial}{\partial t} \hat{\rho}(x, t) \right|_{\text{drift}} = \Gamma \frac{\partial}{\partial x} \hat{\rho}(x, t) \frac{\partial}{\partial x} \int dy \hat{\rho}(y, t) V \left( \frac{|x - y|}{\xi} \right) \quad (2.5)$$

The nucleation of particles is incorporated by adding to the rhs of (2.6) terms of the form

$$\hat{s}(x, t) = \sum_\alpha \delta(x - x_\alpha) \delta(t - t_\alpha) \quad (2.6)$$

where  $t_\alpha$  and  $x_\alpha$  are, respectively, the time and the position at which the  $\alpha$ th nucleation occurs. We assume that the nucleation is a point random (or Poisson random) process and that it is locally specified thoroughly by the nucleation rate  $\hat{I}(x, t)$  such that<sup>(4)</sup>

$$\langle \hat{s}(x_1, t_1) \rangle_{t_1} = \hat{I}(x_1, t_1) \quad (2.7)$$

$$\langle \hat{s}(x_1, t_1) \hat{s}(x_2, t_2) \cdots \hat{s}(x_m, t_m) \rangle_{t_1 c} = \hat{I}(x_1, t_1) \prod_{j=2}^m \delta(x_j - x_1) \delta(t_j - t_1) \quad (2.8)$$

for  $m \geq 2$ . Here  $\langle \cdot \rangle_{t_1}$  and  $\langle \cdot \rangle_{t_1 c}$  represent, respectively, the average and

the corresponding cumulants over random nucleation events *just after*  $t_1$ . In the next section we will use the following identity for  $m \geq 2$ :

$$\langle \hat{s}(x_1, t_1) \cdots \hat{s}(x_m, t_m) \rangle_{t_1 c} = \langle \hat{f}(x_1, t_1) \cdots \hat{f}(x_m, t_m) \rangle_{t_1 c} \quad (2.9)$$

$$\hat{f}(x, t) \equiv \hat{s}(x, t) - \langle \hat{s}(x, t) \rangle_t \quad (2.10)$$

This holds irrespective of the particular probability distribution (see Appendix A).

Next we give the form of the nucleation rate  $\hat{I}(x, t)$  such that it depends on the local density of already existing particles. We assume  $\hat{I}(x, t)$  as follows:

$$\hat{I}(x, t) = \mathcal{F}(M * \hat{\rho}(x, t)) \quad (2.11)$$

where  $M * \hat{\rho}(x, t)$  is the convolution

$$M * \hat{\rho}(x, t) = \int_{-\infty}^{\infty} dy M(x-y) \hat{\rho}(y, t) \quad (2.12)$$

and  $\mathcal{F}(z)$  is a nonnegative function of  $z$ . We assume that  $M(x) = M(-x) \geq 0$  and that  $M(x)$  is characterized by a length  $l_M$  which is much larger than  $\xi$  such that only for  $|x| \lesssim l_M$  is the value of  $M(x)$  appreciable. For convenience we assume that  $M(x)$  is normalized,

$$\int_{-\infty}^{\infty} dx M(x) = 1 \quad (2.13)$$

The  $M * \hat{\rho}(x, t)$  has the meaning of the particle density at time  $t$  averaged over the region of width  $\sim l_M$  around  $x$ , and  $\mathcal{F}(z)$  represents the effect of the preexisting particles *at*  $t$  on the rate of the nearby nucleation event *just after*  $t$ .<sup>(2)</sup> [This means that  $\hat{I}(x, t)$  and  $\hat{s}(x, t)$  are statistically independent.] We do not specify the functional form of  $\mathcal{F}(z)$  as well as  $M(x)$ , but only assume the following properties of  $\mathcal{F}(z)$ : (1)  $\mathcal{F}(z)$  is a monotonically decreasing function of  $z$ , and (2) it vanishes strictly for  $z$  larger than some constant  $\rho_{\text{eq}}$ . This means that as the local density of particles approaches its equilibrium value  $\rho_{\text{eq}}$ , the nucleation of a new particle becomes increasingly unlikely. From the definition of  $l_M$  it is natural to assume that  $\rho_{\text{eq}} \gtrsim l_M^{-1}$ . When the nucleation is of the activation type with the activation energy  $E$ , that is,  $\mathcal{F}(M * \hat{\rho}) \sim e^{-E/T}$ , the above assumption implies that  $E$  diverges as  $M * \hat{\rho}$  approaches  $\rho_{\text{eq}}$  from below. If the divergence of  $E$  as function of  $M * \hat{\rho}$  is stronger than logarithmic, as is usual for the first-order phase transition, one can see that the curvature  $d^2 \mathcal{F}(z)/dz^2$  eventually becomes positive as  $z$  approaches  $\rho_{\text{eq}}$  from below.

Including the nucleation term  $\hat{s}(x, t)$  thus defined, the following equation formally describes the microscopic process of nucleation and drift of particles:

$$\frac{\partial}{\partial t} \hat{\rho}(x, t) = \frac{\partial}{\partial t} \hat{\rho}(x, t) \Big|_{\text{drift}} + \hat{s}(x, t) \tag{2.14}$$

where the first term on the rhs is defined in (2.5). We can construct the master equation equivalent to (2.14). This is shown in Apendix B.

### 3. MACROSCOPIC BEHAVIOR OF THE DENSITY FLUCTUATION IN A SYSTEM WITHOUT DRIFT

In this section we study the model

$$\frac{\partial}{\partial t} \hat{\rho}(x, t) = \hat{s}(x, t) \tag{3.1}$$

where the stochastic properties of  $\hat{s}(x, t)$  are fully described in the last section. For simplicity we specify the initial condition of the particle density to be

$$\hat{\rho}(x, 0) = 0 \tag{3.2}$$

Below we study the macroscopic, or hydrodynamic, behavior of the density fluctuation; the results are summarized in Fig. 1. We introduce the average density of particles  $\rho^*(t)$ , which is the average of  $\hat{\rho}(x, t)$  over all possible random processes:

$$\rho^*(t) = \langle \hat{\rho}(x, t) \rangle \tag{3.3}$$

Here  $\langle \cdot \rangle$  denotes the average over all possible random processes and should not be confused with  $\langle \cdot \rangle_t$ . Because of the uniform initial condition (3.2), it does not depend on  $x$ . As usual,<sup>(6,7)</sup> we decompose  $\hat{\rho}(x, t)$  as

$$\hat{\rho}(x, t) \equiv \rho^*(t) + \delta\hat{\rho}(x, t) \tag{3.4}$$

Accordingly, (3.1) can be formally decomposed as follows:

$$\frac{d}{dt} \rho^*(t) = \langle \mathcal{J}(\rho^*(t) + M * \delta\hat{\rho}(x, t)) \rangle \tag{3.5}$$

$$\begin{aligned} \frac{\partial}{\partial t} \delta\hat{\rho}(x, t) = & \{ \mathcal{J}(\rho^*(t) + M * \delta\hat{\rho}(x, t)) - \langle \mathcal{J}(\rho^*(t) + M * \delta\hat{\rho}(x, t)) \rangle \} \\ & + \hat{f}(x, t) \end{aligned} \tag{3.6}$$

with

$$\hat{f}(x, t) \equiv \hat{s}(x, t) - \mathcal{F}(\rho^*(t) + M * \delta\hat{\rho}(x, t)) \tag{3.7}$$

where we have used (2.7), (2.11), and (2.13). Here  $\hat{f}(x, t)$  is the fluctuation part of the nucleation and obeys, from (2.8) and (2.9),

$$\langle \hat{f}(x, t) \rangle_t = 0 \tag{3.8}$$

$$\begin{aligned} &\langle \hat{f}(x_1, t_1) \cdots \hat{f}(x_m, t_m) \rangle_{t_1 c} \\ &= \mathcal{F}(\rho^*(t_1) + M * \delta\hat{\rho}(x_1, t_1)) \prod_{k=2}^m \delta(x_k - x_1) \delta(t_k - t_1) \end{aligned} \tag{3.9}$$

for  $m \geq 2$ . In the last equation the cumulant  $\langle \cdot \rangle_{t_1 c}$  is taken over the nucleation events just after  $t_1$ .

Our aim is to know from (3.5)–(3.9) the behavior of the macroscopic density fluctuation  $\delta\rho(x, t)$ , which is defined by

$$\delta\rho(x, t) = \frac{1}{\tau} \int_t^{t+\tau} dt' \frac{1}{l} \int_{x-l/2}^{x+l/2} dx' \delta\hat{\rho}(x', t') \tag{3.10}$$

and depends on  $l$  and  $\tau$ , the coarse-graining scales in space and time, respectively, our strategy is (1) to assume

$$\begin{aligned} &|\mathcal{F}(\rho^*(t) + M * \delta\hat{\rho}(x, t)) - \mathcal{F}(\rho^*(t)) - \mathcal{F}'(\rho^*(t)) M * \delta\hat{\rho}(x, t)| \\ &\ll |\mathcal{F}'(\rho^*(t)) M * \delta\hat{\rho}(x, t)| \end{aligned} \tag{3.11}$$

where  $\mathcal{F}'(z) = d\mathcal{F}(z)/dz$ , and then (2) to seek for the condition on  $l$  or  $\tau$  for the consistency of such an assumption. We note here that for any transient process the coarse-graining time scale  $\tau$  should not be larger than the characteristic time scale of the process itself, since otherwise we could not observe the transient process. Therefore in our case we assume that

$$\frac{1}{\rho^*(t)} \frac{d}{dt} \rho^*(t) \lesssim \frac{1}{\tau} \tag{3.12}$$

$$\frac{1}{\mathcal{F}(\rho^*(t))} \left| \frac{d}{dt} \mathcal{F}(\rho^*(t)) \right| \lesssim \frac{1}{\tau} \tag{3.13}$$

Because we have from (3.5) and (3.11)

$$\frac{d}{dt} \rho^*(t) \simeq \mathcal{F}(\rho^*(t)) \tag{3.14}$$

we can reexpress (3.12) and (3.13) as

$$\frac{1}{\rho^*(t)} \lesssim \frac{1}{\mathcal{F}(\rho^*(t))\tau} \tag{3.12'}$$

$$\frac{|\mathcal{F}'(\rho^*(t))|}{\mathcal{F}(\rho^*(t))} \lesssim \frac{1}{\mathcal{F}(\rho^*(t))\tau} \tag{3.13'}$$

Now we expand  $\mathcal{F}(\rho^*(t) + M * \delta\hat{\rho}(x, t))$  in (3.6) in powers of  $M * \delta\hat{\rho}(x, t)$  and average both sides of (3.6) over the coarse-graining scale  $l$  and  $\tau$  as in (3.10). Thus, we have

$$\frac{\partial}{\partial t} \delta\rho(x, t) \simeq \{ \mathcal{F}'(\rho^*(t)) M * \delta\rho(x, t) + \dots \} + f(x, t) \tag{3.15}$$

with

$$f(x, t) \equiv \frac{1}{\tau} \int_t^{t+\tau} dt' \frac{1}{l} \int_{x-l/2}^{x+l/2} dx' \hat{f}(x', t') \tag{3.16}$$

where we have used (3.13), and the ellipsis denotes the contribution from the higher order terms in the expansion of  $\mathcal{F}(\rho^*(t) + M * \delta\hat{\rho}(x, t))$ . Since we have assumed in Section 2 that  $\mathcal{F}'(z) < 0$  for  $0 \leq z < \rho_{eq}$ , the terms in the curly bracket on the rhs of (3.6), and therefore those in (3.15), serve as the negative feedback for the density fluctuation. On the other hand, the assumption (3.13') means that the characteristic decay time of the fluctuation  $\tau_c(\rho^*(t))$ , which is defined from (3.15) by

$$\frac{1}{\tau_c(\rho^*(t))} \equiv |\mathcal{F}'(\rho^*(t))| \tag{3.17}$$

is longer than the coarse-graining time  $\tau$ . Therefore only  $f(x, t)$  is the source of the density fluctuation and it should be balanced with the lhs of (3.15). (In Appendix C we show explicitly such a balance of terms in a simple zero-dimensional model.) In estimating the magnitude of  $f(x, t)$ , we adopt the discretized representation of space and time with their mesh sizes  $l$  and  $\tau$ , respectively, in order to clarify the dependence on these scales. We denote

$$x = Xl \quad X \text{ an integer} \tag{3.18}$$

$$t = T\tau \quad T \text{ an integer} \tag{3.19}$$

Then the cumulants of  $f(x, t)$  can be evaluated from (3.9), (3.11), (3.13), and (3.16) as follows:

$$\langle f(X_1 l, T_1 \tau) \dots f(X_m l, T_m \tau) \rangle_{T_1 c} = (l\tau)^{-m+1} \mathcal{F}(\rho^*(T_1 \tau)) \prod_{j=2}^m \delta_{X_j X_1} \delta_{T_j T_1} \tag{3.20}$$

where  $m \geq 2$ , and  $\delta_{ij}$  is the Kronecker delta function. The cumulant  $\langle \cdot \rangle_{T_1}$  is taken over the nucleation events between  $T_1\tau$  and  $(T_1+1)\tau$ . It is  $\mathcal{J}(\rho^*(t))$  that should be compared with  $f(x, t)$ , since these lead, respectively, to the random and the systematic variations of the macroscopic density. From (3.20) we obtain

$$\left\langle \frac{f(X_1 l, T_1 \tau)}{\mathcal{J}(\rho^*(T_1 \tau))} \cdots \frac{f(X_m l, T_m \tau)}{\mathcal{J}(\rho^*(T_m \tau))} \right\rangle_{T_1 c} = \{\mathcal{J}(\rho^*(T_1 \tau)) l \tau\}^{-m+1} \prod_{j=2}^m \delta_{x_j x_1} \delta_{T_j T_1} \quad (3.21)$$

Therefore, if

$$\mathcal{J}(\rho^*(t)) l \tau \gg 1 \quad (3.22)$$

the cumulants with  $m \geq 3$  are negligible and

$$\frac{f(x, t)}{\mathcal{J}(\rho^*(t))} \sim \{\mathcal{J}(\rho^*(t)) l \tau\}^{-1/2} \ll 1 \quad (3.23)$$

can be regarded as the Gaussian noise, which is uncorrelated or white on scales larger than  $l$  and  $\tau$ . From (3.22), (3.12'), and (3.13') we obtain the conditions

$$\frac{1}{\rho^*(t)} \ll l \quad (3.24)$$

$$\frac{|\mathcal{J}'(\rho^*(t))|}{\mathcal{J}(\rho^*(t))} \ll l \quad (3.25)$$

If these are satisfied, then we can make  $f(x, t)$  an uncorrelated Gaussian noise by properly choosing  $\tau$  within the bounds given by (3.12') and (3.13'). In Figs. 1a and 1b we show schematically (3.24) and (3.25) in the  $(\rho^*, l)$  plane. In order to see the behavior of  $\delta\rho(x, t)$  from (3.15), we classify the process into several stages and study each of them separately.

I.  $\rho^*(t) \ll l_M^{-1}$  (the early stage): In this stage it is quite unlikely that the nucleation occurs at a distance less than  $\sim 2l_M$  from any one of the preexisting particles. Therefore, the nucleation rate  $\mathcal{J}(\rho^*(t) + M * \delta\rho(x, t))$  should be practically equal to  $\mathcal{J}(0)$ . In this sense (3.11) is valid and we have from (3.14) and (3.15)

$$\frac{d}{dt} \rho^*(t) \simeq \mathcal{J}(0) \quad (3.26)$$

$$\frac{\partial}{\partial t} \delta\rho(x, t) \simeq f(x, t) \quad (3.27)$$



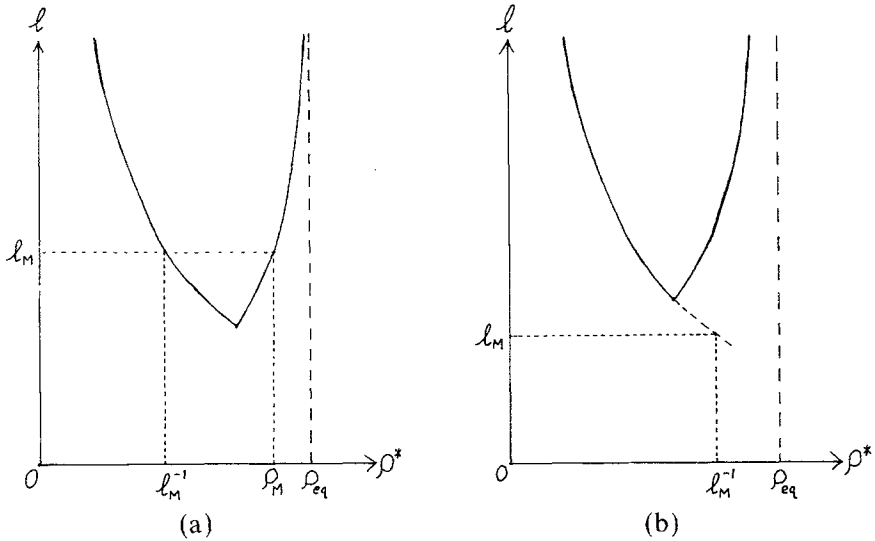


Fig. 1. Schematic diagrams indicating the regimes in which a hydrodynamic description may be valid. The thick line in each figure represents the larger of the lhs of (3.24) and the lhs of (3.25). Therefore if  $l$  is well above this line, both (3.24) and (3.25) are satisfied. (a, b) Possible cases according to whether there does (does not) exist a time domain with  $l_M^{-1} < \rho^*(t) < \rho_M$  in which the thick line is well below the horizontal line  $l = l_M$ .

If we choose the coarse-graining scale  $l$  so that it satisfies (3.24) and (3.25), the random source  $f(x, t)$ , which can be regarded as the Gaussian noise [cf. (3.21)], is characterized by the following variance:

$$\langle f(Xl, T\tau) f(X'l, T'\tau) \rangle_T = \frac{\mathcal{F}(0)}{l\tau} \delta_{XX'} \delta_{TT'} \tag{3.28}$$

in the discretized representation.

For  $\rho^*(t) \gtrsim l_M^{-1}$  we have to distinguish the following two cases:

Case a.  $l_M \gg |\mathcal{F}'(l_M^{-1})|/\mathcal{F}(l_M^{-1})$  (see Fig. 1a)

Case b.  $l_M \lesssim |\mathcal{F}'(l_M^{-1})|/\mathcal{F}(l_M^{-1})$  (see Fig. 1b)

In case a there is the following stage.

II.  $l_M^{-1} \lesssim \rho^*(t) \lesssim \rho_M$  with  $|\mathcal{F}'(\rho_M)|/\mathcal{F}(\rho_M) \equiv l_M$  (the intermediate stage for case a only): In this stage we may choose  $l$  (as in fact we do) such that

$$\text{Max} \left( \frac{1}{\rho^*(t)}, \frac{|\mathcal{F}'(\rho^*(t))|}{\mathcal{F}(\rho^*(t))} \right) \ll l \lesssim l_M \tag{3.29}$$

With this choice we can replace  $M * \delta\hat{\rho}(x, t)$  in (3.5)–(3.11) by  $M * \delta\rho(x, t)$ , since  $M * \delta\hat{\rho}(x, t)$  is essentially the macroscopic density averaged over  $\sim l_M$  ( $\gtrsim l$ ). Therefore, from (3.5), (3.6), (3.10), and (3.16) we have for  $l \lesssim l_M$

$$\frac{d}{dt} \rho^*(t) = \langle \mathcal{I}(\rho^*(t) + M * \delta\rho(x, t)) \rangle \tag{3.30}$$

$$\frac{\partial}{\partial t} \delta\rho(x, t) = \mathcal{I}(\rho^*(t) + M * \delta\rho(x, t)) - \langle \mathcal{I}(\rho^*(t) + M * \delta\rho(x, t)) \rangle + f(x, t) \tag{3.31}$$

Because of the left inequality of (3.29), the random source  $f(x, t)$  can be approximated by the Gaussian noise with

$$\langle f(Xl, T\tau) f(X'l, T'\tau) \rangle_{T\tau} = \frac{\mathcal{I}(\rho^*(T\tau) + M * \delta\rho(Xl, T\tau))}{l\tau} \delta_{XX'} \delta_{TT'} \tag{3.32}$$

This is small compared with  $\mathcal{I}(\rho^*(t) + M * \delta\rho(x, t))$  by the order of

$$[\mathcal{I}(\rho^*(t) + M * \delta\rho(x, t)) l\tau]^{-1/2}$$

The assumption (3.11) is now written as follows:

$$\begin{aligned} & |\mathcal{I}(\rho^*(t) + M * \delta\rho(x, t)) - \mathcal{I}(\rho^*(t)) - \mathcal{I}'(\rho^*(t)) M * \delta\rho(x, t)| \\ & \ll |\mathcal{I}'(\rho^*(t)) M * \delta\rho(x, t)| \end{aligned} \tag{3.11'}$$

By solving (3.30)–(3.32) under this assumption, we obtain, neglecting the initial transient terms,

$$\delta\rho(x, t) = \int dx e^{ikx} \delta\tilde{\rho}_k(t) \tag{3.33}$$

$$\delta\tilde{\rho}_k(t) = \int^t ds \exp \left\{ \tilde{M}_k \int_s^t du \mathcal{I}'(\rho^*(u)) \right\} \tilde{f}_k(s) \tag{3.34}$$

where  $\tilde{M}_k$  and  $\tilde{f}_k$  are the spatial Fourier components of  $M(x)$  and  $f(x, s)$ , respectively, which are defined in the same manner as  $\delta\tilde{\rho}_k(t)$  in (3.33). Note that these Fourier components are cut off for  $|k| \gtrsim l^{-1}$  ( $\gtrsim l_M^{-1}$ ). The structure function  $S_k(t)$ , which is defined by

$$\langle \delta\tilde{\rho}_k(t) \delta\tilde{\rho}_{k'}(t) \rangle = 2\pi\delta(k + k') S_k(t) \tag{3.35}$$

is given from (3.32) and (3.34) as

$$S_k(t) = \int^t ds \mathcal{I}(\rho^*(s)) \exp \left\{ [\tilde{M}_k + \tilde{M}_{-k}] \int_s^t du \mathcal{I}'(\rho^*(u)) \right\} \tag{3.36}$$

or, by using (3.14),

$$S_k(t) = \int^{\rho^*(t)} d\rho \left\{ \frac{\mathcal{F}(\rho^*(t))}{\mathcal{F}(\rho)} \right\}^{\tilde{M}_k + \tilde{M}_{-k}} \tag{3.37}$$

$S_k(t)$  is also cut off for  $|k| > l^{-1}$ . The magnitude of  $\delta\rho(x, t)$  can be evaluated by the following formula:

$$\langle \delta\rho(x, t)^2 \rangle = \int \frac{dk}{2\pi} S_k(t) \tag{3.38}$$

If  $\tilde{M}_k + \tilde{M}_{-k}$  is positive, as we assume [see Appendix D, where  $S_k(t)$  is evaluated for a concrete model of  $\mathcal{F}(z)$  and  $M(x)$ ], we find

$$S_k(t) \lesssim \rho^*(t) \theta(l^{-1} - |k|) \tag{3.39}$$

because  $\mathcal{F}(\rho^*(t))/\mathcal{F}(\rho) \leq 1$  if  $\rho \leq \rho^*(t)$  and there is an upper cutoff at  $|k| \sim l^{-1}$ , where  $\theta(z)$  is the usual unit step function. Therefore we have

$$\frac{\langle \delta\rho(x, t)^2 \rangle}{\rho^*(t)^2} \lesssim \frac{1}{\rho^*(t)l} \ll 1 \tag{3.40}$$

This estimation is very natural, since for completely random nucleation the left inequality in (3.40) should become the equality, and in our model the density fluctuation is suppressed for  $|k| < l_M^{-1}$  in comparison to the completely random case. If we substitute (3.37) and (3.38) into (3.11'), we obtain another constraint on  $l$ , which is not shown in Figs. 1a and 1b. We expect from (3.40) that this constraint also gives the lower bound of  $l$ . It is clear that the results (3.11')–(3.40) hold also for  $l > l_M$ , since once the linearization approximation for (3.31) is guaranteed for  $l \lesssim l_M$  the further coarse-graining over  $l > l_M$  merely leads to the red shift of the wavenumber upper cutoff.

We note that the expressions (3.34)–(3.37) are also valid for  $l \gg \rho^*(t)^{-1}$  in the initial stage I [see (3.26) and (3.27)] if the intermediate stage exists. This is because in this case  $l_M \gg |\mathcal{F}'(l_M^{-1})|/\mathcal{F}(l_M^{-1})$  holds, which in turn means that

$$\mathcal{F}(\rho^*(t)) \simeq \mathcal{F}(0) \quad \text{for } 0 \leq \rho^*(t) \lesssim l_M^{-1} \tag{3.41}$$

In Appendix E we develop the path integral formalism for the evolution of  $\delta\rho(x, t)$ , which is valid in the intermediate (and initial) stages.

III.  $\rho^*(t) \gtrsim \rho_M$  for case a and  $\rho^*(t) \gtrsim l_M^{-1}$  for case b (the final stage): In this stage the conditions (3.24) and (3.25) imply  $l \gg l_M$ . Therefore, if we choose  $l \sim l_M$ , Eqs. (3.30) and (3.31) are valid, but the noise  $f(x, t)$  can no

longer be regarded as a white Gaussian noise. In order to obtain  $\rho^*(t)$  or  $\langle \delta\rho(x, t) \delta\rho(x', t) \rangle$ , etc., in such choice of  $l$ , we must go back to (3.5)–(3.9) and study them kinematically. The result should reflect the nonlinearity of  $\mathcal{J}(z)$  as well as the detailed form of  $M(x)$  [in fact, the condition (3.25) might be altered due to such nonlinearity]. On the other hand, if we choose  $l$  such that (3.24) and (3.25) (or its improved form) are satisfied,  $f(x, t)$  and  $\delta\rho(x, t)$  become the Gaussian random variables whose variances are obtained only from the kinetic theory mentioned above. The reasons are as follows: (1) There is no correlation between the nucleation events occurring at a distance greater than  $l_M$ , which is the definition of  $l_M$  (see also Appendix D), and (2)  $f(x, t)$  and  $\delta\rho(x, t)$  are the averages over  $\sim l/l_M (\gg 1)$  of independent variables representing each correlation region of the size  $\sim l_M$ . Then, from the central limit theorem,  $f(x, t)$  and  $\delta\rho(x, t)$  become the Gaussian random variables whose variances are both  $\sim l_M/l (\ll 1)$  times those to each correlation region.

In Fig. 2 we show  $S_k(t)$  obtained by the computer simulation in which  $\mathcal{J}(z)$  and  $M(x)$  are defined by (D.1) and (D.2). The simulation method is

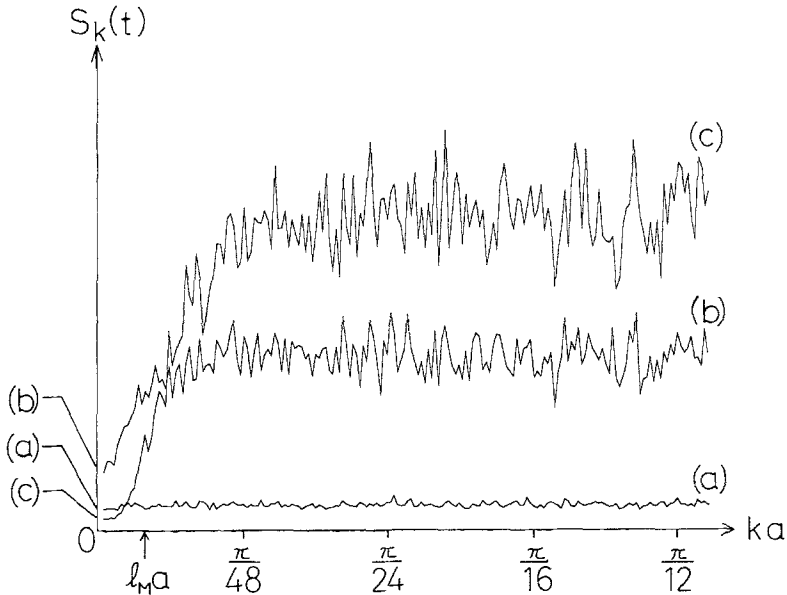


Fig. 2. The structure function  $S_k(t)$  obtained from the computer simulation (see the text) as a function of  $ka$  at various values of time, where  $a$  is the unit length of our simulation with which the space was discretized. The parameters used in the model [see (D.1) and (D.2)] are  $\rho_{\text{eq}} a = \rho_0 a = 0.05$  and  $l_M a^{-1} = 50$ . The system size is  $2048a$  with the periodic boundary condition, and  $S_k(t)$  is the average taken over 100 runs. The time expressed in the dimensionless form  $\mathcal{J}_0 a t$  and the average density of particles  $\rho^*$  in units of  $\rho_{\text{eq}}$  are: (a)  $(\mathcal{J}_0 a t, \rho^*/\rho_{\text{eq}}) = (4 \times 10^{-3}, 0.1)$ , (b)  $(4 \times 10^{-2}, 0.5)$ , and (c)  $(1, 0.97)$ .

the so-called the event-waiting method, in which we utilize the random-number generator to determine not only the positions, but also the times at which the nucleation events occur. From this figure we see clearly that the small- $|k|$  components of the density fluctuation are suppressed in the later stages. We see also that the characteristic wavenumber  $k = k_c(t)$  above which the density fluctuations are not suppressed is an increasing function of time. This means that the width of the correlation hole in the density correlation function (see Appendix D) decreases with time, which coincides with the result of Appendix D.

From the general point of view the intermediate stage in case a is rather specific, since in that stage we could perform the coarse-graining without considering the kinetic correlation among particles. This is because in our model the density dependence of the nucleation rate is mean-field-like over the range  $\sim l_M$ , inside of which the nucleation events are completely random (i.e., Poissonian) at least for a short time interval. Such a mean-field-like nature of our model might be shared with the charged particle system,<sup>(7)</sup> where the soft collision is important. In the C-IC transition an array of already existing discommensurations can be deformed when a nucleation occurs within the array. The long-range nature of the deformation field<sup>(8)</sup> will be important, which might have something in common with the present analysis.

#### 4. NONLINEAR DRIFT OF PARTICLES

In this section we study the model without nucleation, which is defined by (2.3), or

$$\frac{d}{dt} x_n = -\frac{\Gamma}{\xi} \left\{ V' \left( \frac{x_n - x_{n-1}}{\xi} \right) - V' \left( \frac{x_{n+1} - x_n}{\xi} \right) \right\} \quad (4.1)$$

and discuss the effect of nucleation on the motion of preexisting particles. Since we assume that typically  $|x_n - x_{n-1}|/\xi$  is much greater than unity, which is usual near the C-IC transition, we have to distinguish the following two cases:

1. The interparticle distance  $\Delta_n$  defined by

$$\Delta_n = x_{n+1} - x_n \quad (4.2)$$

varies rapidly with  $n$ , that is,

$$|\Delta_{n-1} - \Delta_n| \gtrsim \xi \quad (4.3)$$

2. The variation of  $\Delta_n$  is small, that is,

$$|\Delta_{n-1} - \Delta_n| \ll \xi \quad (4.4)$$

In general these two cases will be realized at the same time in different regions of the system.

First we consider case 1 and suppose that the nucleation also occurs occasionally. In this case it is impossible to carry out the coarse-graining procedure on the rhs of (4.1) in a simple way because the "collision" of particles, which is the random nucleation in our model, is not frequent enough to establish the local equilibrium (or steadiness) within the coarse-graining time interval  $\tau$ . In fact, the number of nucleation events per particle is  $\sim \mathcal{J}(\rho^*(t))\tau/\rho^*(t)$  for the time interval  $(t, t + \tau)$ , while from (3.12') we should choose  $\mathcal{J}(\rho^*(t))\tau/\rho^*(t) \lesssim 1$  so that we can observe the transition period. In the above argument we have neglected the secondary or higher order collisions between particles. We expect that they are not so efficient for establishing the local steady-state condition. If the (transient) process involves both nucleation and annihilation of particles, the local equilibrium condition can be fulfilled as in the chemical reaction process.

Next we consider case 2. It has been shown<sup>(3)</sup> that we can regard  $\Delta_n^{-1}$  as the slowly varying density on the real space,

$$\rho(x, t) \simeq \Delta_n(t)^{-1} \quad \text{for } x \simeq x_n(t) \quad (4.5)$$

and that (4.1) can be converted via expression (2.5) into the form

$$\frac{\partial}{\partial t} \rho(x, t) = -\frac{\Gamma}{\xi} \frac{\partial^2}{\partial x^2} V' \left( \frac{1}{\rho(x, t)\xi} \right) \quad (4.6)$$

[In Ref. 3 only the case of  $V(z) = e^{-z}$  was studied. However, the extension for the general symmetric interaction  $V(z)$  is straightforward.] Alternatively, we may regard  $\Delta_n$  as the slowly varying field in the space of the variable  $n$ ,

$$u(n, t) \simeq \Delta_n(t) \quad (4.7)$$

where  $n$  on the lhs can be extended to assume real, continuous values. Then (4.1) becomes<sup>(9)</sup>

$$\frac{\partial}{\partial t} u(n, t) = \frac{\Gamma}{\xi} \frac{\partial^2}{\partial n^2} V' \left( \frac{u(n, t)}{\xi} \right) \quad (4.8)$$

Equations (4.6) and (4.8) give the same level of description in case 2 and the condition (4.4) is converted into

$$\left| \frac{\partial}{\partial x} \rho(x, t) \right| \ll \rho(x, t)^3 \xi \quad (4.9)$$

or

$$\left| \frac{\partial}{\partial n} u(n, t) \right| \ll \xi \tag{4.10}$$

Since our interest is in the case of  $\Delta_n(t)/\xi \simeq 1/\rho(x, t)\xi \gg 1$ , the above condition will severely limit the applicability of (4.6) and (4.8) in the presence of nucleation. In the nonlinear diffusion equations (4.6) and (4.8) the nonlinearity of the repulsive force  $V'(z)$  leads to the preferential diffusion of the conserved quantities  $\rho(x, t)$  and  $u(n, t)$  where the particles are crowded, that is, where  $\rho(x, t)$  and  $[u(n, t)]^{-1}$  are large.

Other aspects of the nonlinearity of  $V'(z)$  are seen when the array of particles can be divided into a set of domains within each of which the continuum description (case 2 above) is valid. Two typical examples of such situations are discussed below.

(a) A cluster of particles. Suppose that  $N$  particles are initially placed in the following manner:

$$x_n(0) = n\Delta_0, \quad n = 1, 2, \dots, N \tag{4.11}$$

The evolution in the case of  $N = \infty$  has been previously studied.<sup>(9)</sup> There it was shown that (1) the continuum approximation (4.8) is practically valid up to the boundary of the cluster, though (4.4) is not satisfied for  $n = 1$ , (2) the boundary condition is effectively given as

$$u(0, t) = \infty \tag{4.12}$$

or

$$V'(u(0, t)) = 0 \tag{4.13}$$

and (3) the drift of the particles leads to the smearing of the cluster boundary and obeys the following scaling law:

$$u(n, t) \simeq \tilde{u}(nt^{-1/2}), \quad n > 0 \tag{4.14}$$

$$x_n(t) \simeq t^{1/2} \tilde{x}(nt^{-1/2}), \quad n = 1, 2, \dots \tag{4.15}$$

where the scaling functions  $\tilde{u}(z)$  and  $\tilde{x}(z)$  depend functionally on  $V'(z)$  as well as  $\Delta_0$ . Relations (4.14) and (4.15) show that the width of the smeared boundary grows as  $t^{1/2}$  both in  $x$  space and in  $n$  space.

Such smearing of the boundary should also occur in the present case of  $N < \infty$  until the two smeared boundaries, which have been localized initially at  $n = 1$  and  $N$ , overlap with each other in the middle of the cluster.

This mode of relaxation may be called the boundary mode. The duration of this mode  $t_b$  will depend on  $N$  as

$$t_b \propto N^2 \quad (4.16)$$

For  $t \gtrsim t_b$  another mode of relaxation occurs. Since there is no universal scaling law in this mode, we will study the following class of interparticle force:

$$\tilde{F}(z) \equiv -V'(z) = e^{-(z+b)} \quad (4.17)$$

where  $b$  is some constant. If we introduce  $F(n, t)$  by

$$F(n, t) = \tilde{F}(u(n, t)/\xi) \quad (4.18)$$

then we have from (4.8), (4.17), and (4.18)

$$\frac{\partial}{\partial t} F = \gamma F \frac{\partial^2}{\partial n^2} F, \quad \gamma = \xi^{-2} \Gamma \quad (4.19)$$

with the boundary condition [see (4.13)]

$$F(0, t) = F(N, t) = 0 \quad (4.20)$$

We can find the following special solution of (4.19) and (4.20), which describes the smearing of the cluster itself, which we call the bulk mode:

$$F_b(n, t) = \frac{1}{2\gamma t} n(N-n) \quad (4.21)$$

The cluster size  $L(t) \equiv x_N(t) - x_1(t)$  can be approximated as

$$L(t) = \sum_{n=1}^{N-1} A_n(t) \quad (4.22)$$

$$\simeq 2 \int_0^{N/2} dn u(n, t) \quad (4.23)$$

The behavior of  $L(t)$  for  $t \gg t_b$  is given from (4.17), (4.18), (4.21), and (4.23) as follows:

$$L(t) \sim \xi N \log t \quad (4.24)$$

The rate of growth of  $L(t)$  is slowed compared with the boundary mode. In Appendix F we argue that the solution (4.21) will be an attractor of solutions, or that the small deviation of  $F(n, t)$  from  $F_b(n, t)$  asymptotically vanishes.



If there are many clusters containing various numbers of particles and separated from each other by various distances, their evolution will involve not only the two modes of cluster relaxation, i.e., the boundary mode and the bulk mode, but also the collision of these clusters, all of which depend on the size of the clusters and the intercluster distances.

(b) A particle added to an otherwise periodic array of particles. We consider the evolution of (4.1) from the following initial configuration:

$$x_0(0) = 0 \tag{4.25}$$

$$x_n(0) = \begin{cases} (n - \frac{1}{2}) \Delta_\infty, & n = 1, 2, \dots \\ -(|n| - \frac{1}{2}) \Delta_\infty, & n = -1, -2, \dots \end{cases} \tag{4.26}$$

$$\tag{4.27}$$

We expect that, in the final stage of the nucleation-plus-drift process, a configuration of the particles such as that given by (4.25)–(4.27) will be found (see the last paragraph in this section). The motion of particles from the initial condition (4.25)–(4.27) is shown in Fig. 3, which was obtained numerically for  $\Delta_\infty/\xi = 10$ . We can see that the shocklike fronts propagate toward opposite directions. The fronts separate the high-density region (between the fronts), and the low-density region (the outside), in each of which the spatiotemporal variation of interparticle distances is weak. These shocklike fronts originate in the strong nonlinearity of the interparticle repulsion under the variation of interparticle distance  $\Delta_n = x_{n+1} - x_n$  over

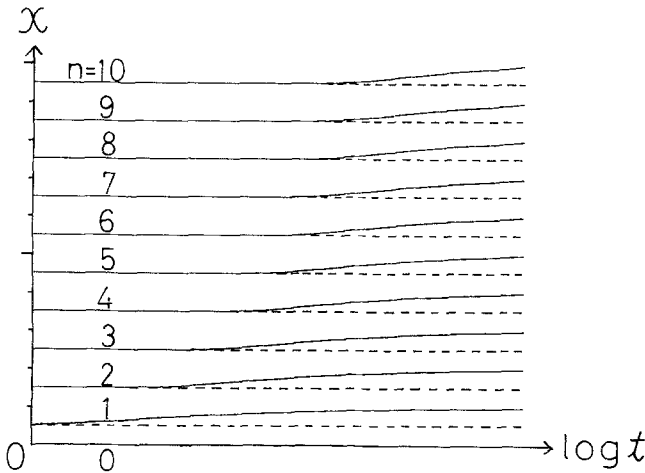


Fig. 3. (—) The trajectories of particles  $x = x_n(t)$  that obey the equation of motion (4.1) with the initial conditions (4.25)–(4.27). The abscissa is the logarithm of time. The trajectories with  $n < 0$ , which are not shown in the figure, are symmetric with respect to the abscissa,  $x_{-n}(t) = -x_n(t)$ . (---) The initial positions of each particle.

a range much greater than  $\xi$ . In fact, when the front is at the  $n$ th particle with  $n \geq 1$ , we have

$$\left| V' \left( \frac{x_n(t) - x_{n-1}(t)}{\xi} \right) \right| \gg \left| V' \left( \frac{x_{n+1}(t) - x_n(t)}{\xi} \right) \right| \tag{4.28}$$

for

$$D_n(t) \equiv \{x_{n+1}(t) - x_n(t)\} - \{x_n(t) - x_{n-1}(t)\} \gg \xi \tag{4.29}$$

(for example, consider the case of  $-V'(z) \propto e^{-z}$  or  $e^{-z^2}$ ). Then the motion of the  $n$ th particle is governed mostly by the repulsive force coming from the side of the high-density region, and therefore the velocity of the  $n$ th particle [and the  $(-n)$ th particle by symmetry] is the greatest. The only exception to (4.28) occurs when  $x_n(t)$  reaches the moving midpoint of its two neighbors, that is, when

$$0 < D_n(t) \lesssim \xi \tag{4.30}$$

is satisfied. At this moment, which we denote as  $t \simeq t_{n+1}$ , the shocklike front is shifted from  $x_n(t)$  to  $x_{n+1}(t)$  and the inequality (4.28) may again hold, except that now  $n$  is replaced by  $n + 1$ , that is,

$$D_{n+1}(t_{n+1}) \gg \xi \tag{4.31}$$

In this way the shocklike front propagates in  $n$  space as well as in the  $x$  space. However, the quantity  $D_n(t_n)$  decreases as the shocklike front propagates in  $n$  space (i.e., the shock strength weakens), that is,

$$\dots > D_n(t_n) > D_{n+1}(t_{n+1}) > \dots \tag{4.32}$$

Eventually, at a certain  $n$ , which we denote  $n_c$ , we will have

$$D_{n_c}(t_{n_c}) \sim \xi \tag{4.33}$$

For  $t > t_{n_c}$  the shocklike front no longer exists and the continuum approximation (4.6) or (4.7) becomes valid in the entire  $x$  or  $n$  space.

The quantity  $x_{n_c} t_{n_c}$  is the characteristic space-time volume over which an added particle strongly distorts the otherwise almost periodic array of particles. It depends on the period  $\Delta_\infty$  of the array or the density  $\rho^* \equiv 1/\Delta_\infty$ . Therefore, if the addition of a particle is governed by the nucleation rate  $\mathcal{F}(\rho^*)$ , then

$$\theta(\rho^*) \equiv \mathcal{F}(\rho^*) x_{n_c} t_{n_c} \tag{4.34}$$

measures the degree of randomness in the array of particles. In particular, if  $\theta(\rho^*) \ll 1$ , the continuum approximation (4.6) for the particle drift is valid on the whole. Since  $\mathcal{J}(\rho^*)$  vanishes as  $\rho^* \rightarrow \rho_{\text{eq}}$  while  $x_{nc}$  and  $t_{nc}$  remain finite, this inequality characterizes the final stage of the drift and nucleation process.

## 5. SUMMARY

The main features of the nucleation and/or drift process in our model are summarized as follows:

When we consider only the nucleation, the process can be divided into two or three stages from the statistical viewpoint. The first is the gaslike stage when the correlation between the nucleation and the distribution of preexisting particles are negligible. The second, when it exists, is the stage where such correlation is not negligible, but is not yet so strong as to severely suppress the nucleation. The third stage is the final stage, when the density of particles is almost saturated and therefore further nucleation must reflect the strong nonlinearity in the nucleation rate as a function of the particle density.

If we are to include the drift of particles caused by the weak and non-linear repulsive interaction, the distribution of particles is found to have the following features. The local equilibrium or steadiness of the particle distribution is not easily established, which makes it impossible to obtain the hydrodynamic expression for the drift term in the evolution equation. This is because in transient processes the observation time cannot be made sufficiently long for a hydrodynamic description without allowing the macroscopic state itself to change. On the other hand, the nucleation in the final stage of the process may be regarded as the sudden addition of a particle in an otherwise periodic array of particles. Then the drift of particles following the nucleation can be described as the propagation of shock fronts, which subsequently disappear eventually. The statistics of the particle density depends on the number of such shock fronts generated in a given space-time volume.

In a future study of transient processes involving the formation of various patterns one of the important problems will be to clarify how and to what degree the past history of the process is recorded in the present irregular patterns.

## APPENDIX A

We derive here the identity

$$\langle (X - \langle X \rangle)^n \rangle_c = \langle X^n \rangle_c, \quad n \geq 2 \quad (\text{A.1})$$

where  $\langle \cdot \rangle$  and  $\langle \cdot \rangle_c$  are the average and the corresponding cumulant over an arbitrarily chosen probability distribution. The extension to the multi-variable case is straightforward, which leads to (2.9) in the text. We start from the formal identity

$$\langle e^{aX+bY} - 1 \rangle_c = \log \langle e^{aX+bY} \rangle \quad (\text{A.2})$$

which defines the cumulants order by order in both  $a$  and  $b$ . We see that if the variable  $Y$  is in fact a constant  $Y_0$ , then we have

$$\langle e^{aX+bY} - 1 \rangle_c = \log(\langle e^{aX} \rangle e^{bY}) = \langle e^{aX} - 1 \rangle_c + bY_0 \quad (\text{A.3})$$

By choosing  $Y_0 = -\langle X \rangle$  and setting  $a = b$ , we obtain

$$\langle \exp[a(X - \langle X \rangle)] - 1 \rangle_c = \langle \exp(aX) - 1 \rangle_c - a\langle X \rangle \quad (\text{A.4})$$

This in each order of  $a$  leads to the identity (A.1).

## APPENDIX B

The master equation equivalent to (2.14) is given as follows:

$$\begin{aligned} \frac{\partial}{\partial t} P_N(\{x_m\}_N, t) = & - \sum_{n=1}^N \frac{\partial}{\partial x_n} \left\{ \left( \frac{dx_n}{dt} \right)_N P_N(\{x_m\}_N, t) \right\} \\ & + (1 - \delta_{N,0}) \sum_{n=1}^N I(\{x_m\}_N - x_n; x_n) P_{N-1}(\{x_m\}_N - x_n, t) \\ & - \int dy I(\{x_m\}_N; y) P_N(\{x_m\}_N, t) \end{aligned} \quad (\text{B.1})$$

where  $P_N(\{x_m\}_N, t)$  is the probability that at time  $t$  there are  $N$  particles with their positions at  $\{x_m\}_N = \{x_1, x_2, \dots, x_N\}$ , and  $(dx_n/dt)_N$  stands for the rhs of (2.2) in the text. The last two terms on the rhs describe the nucleation of a particle, where the nucleation rate  $I(\cdot; y)$  is related to  $\hat{I}(y, \cdot) = \mathcal{J}(M * \hat{\rho}(y, \cdot))$  as follows:

$$I(\{x_m\}_N; y) = \mathcal{J}(M * \hat{\rho}(y, \cdot)) \quad (\text{B.2})$$

$$\hat{\rho}(y, \cdot) = \sum_{m=1}^N \delta(y - x_m) \quad (\text{B.3})$$

The notation  $\{x_m\}_N - x_n$  denotes  $\{x_1, \dots, x_{n-1}, x_{n+1}, \dots, x_N\}$ , and  $\delta_{N,0}$  is unity if  $n = 0$  and zero otherwise.

The equivalence between (2.14) and (B.1) can be seen by comparing the evolution equations of the (multi-) particle density distribution function. As an illustration, we consider the two-particle distribution function defined by

$$\rho_2(x, y; t) = \sum_{N=0}^{\infty} \int_N P_N(\{x_m\}_N, t) \sum_{k=1}^N \delta(x-x_k) \sum_{l=1}^N \delta(y-x_l) \quad (B.4)$$

where  $\int_N$  denotes  $(N!)^{-1} \int dx_1 \int dx_2 \cdots \int dx_N$ . Operating with  $\partial/\partial t$  on (B.4) and using (B.1), we obtain after some algebra

$$\begin{aligned} & \frac{\partial}{\partial t} \rho_2(x, y; t) \\ &= \left\langle \left\langle \left\{ -\frac{\partial}{\partial x} \sum_{k=1}^N \delta(x-x_k) \left( \frac{dx_k}{dt} \right)_N + I(\{x_m\}_N; x) \right\} \sum_{l=1}^N \delta(y-x_l) \right\rangle \right\rangle_t \\ &+ \left\langle \left\langle \left\{ -\frac{\partial}{\partial y} \sum_{k=1}^N \delta(y-x_k) \left( \frac{dx_k}{dt} \right)_N + I(\{x_m\}_N; y) \right\} \sum_{l=1}^N \delta(x-x_l) \right\rangle \right\rangle_t \\ &+ \left\langle \left\langle I(\{x_m\}_N; x) \right\rangle \right\rangle_t \delta(x-y) \end{aligned} \quad (B.5)$$

where we have defined

$$\left\langle \left\langle \mathcal{O}_N(\{x_m\}_N; \cdot) \right\rangle \right\rangle_t = \sum_{N=0}^{\infty} \int_N P_N(\{x_m\}_N, t) \mathcal{O}_N(\{x_m\}_N; \cdot) \quad (B.6)$$

On the other hand, the two-particle density distribution can be expressed as follows:

$$\rho_2(x, y; t) = \langle \hat{\rho}(x, t) \hat{\rho}(y, t) \rangle \quad (B.7)$$

where  $\langle \cdot \rangle$  denotes the average over all possible random processes starting from a given initial condition. [We will use the same notation  $\langle \cdot \rangle$  as in (2.7) unless confusion could arise.] By expressing  $\partial \rho(x, t)/\partial t|_{\text{drift}}$  in (2.14) as  $-\partial \hat{J}(x, t)/\partial x$  [see (2.5)], we have

$$\begin{aligned} & \frac{\partial}{\partial t} \langle \hat{\rho}(x, t) \hat{\rho}(y, t) \rangle \\ &= \lim_{\varepsilon \rightarrow 0} \frac{1}{\varepsilon} \{ \langle [\hat{\rho}(x, t+\varepsilon) - \hat{\rho}(x, t)] \hat{\rho}(y, t) \rangle \\ &+ \langle \hat{\rho}(x, t) [\hat{\rho}(y, t+\varepsilon) - \hat{\rho}(y, t)] \rangle \\ &+ \langle [\hat{\rho}(x, t+\varepsilon) - \hat{\rho}(x, t)] [\hat{\rho}(y, t+\varepsilon) - \hat{\rho}(y, t)] \rangle \} \end{aligned}$$

$$\begin{aligned}
&= \left\langle \left\{ -\frac{\partial}{\partial x} \hat{J}(x, t) + \hat{I}(x, t) \right\} \hat{\rho}(y, t) \right\rangle \\
&\quad + \left\langle \hat{\rho}(x, t) \left\{ -\frac{\partial}{\partial y} \hat{J}(y, t) + \hat{I}(y, t) \right\} \right\rangle \\
&\quad + \lim_{\varepsilon \rightarrow 0} \frac{1}{\varepsilon} \left\{ \int_t^{t+\varepsilon} dt_1 \int_t^{t+\varepsilon} dt_2 \langle \langle \hat{s}(x, t_1) \hat{s}(y, t_2) \rangle_{n_c} \rangle + \mathcal{O}(\varepsilon^2) \right\} \\
&= \left\langle \left\{ -\frac{\partial}{\partial x} \hat{J}(x, t) + \hat{I}(x, t) \right\} \hat{\rho}(y, t) \right\rangle \\
&\quad + \left\langle \hat{\rho}(y, t) \left\{ -\frac{\partial}{\partial y} \hat{J}(y, t) + \hat{I}(y, t) \right\} \right\rangle \\
&\quad + \langle \hat{I}(x, t) \rangle \delta(x - y) \tag{B.8}
\end{aligned}$$

where  $\langle \langle \hat{s}\hat{s} \rangle_{n_c} \rangle$  on the rhs of the second equality means that the cumulant of  $\hat{s}(x, t_1) \hat{s}(y, t_2)$  should be taken first over the random nucleation events during the infinitesimal time interval  $(t_1, t_1 + \varepsilon)$  with  $\hat{\rho}(\cdot, t_1)$  fixed and then over all possible random processes up to the time  $t_1$ . The complete correspondence between (B.5) and (B.8) illustrates the equivalence between (B.1) and (2.14) in the text.

## APPENDIX C

We consider the Langevin equation

$$\frac{d}{dt} X(t) = -\frac{1}{\tau_c} X(t) + f(t) \tag{C.1}$$

where  $\tau_c$  is the characteristic decay time of the ‘‘fluctuation’’  $X(t)$  and  $f(t)$  is the white Gaussian noise whose mean  $\langle f(t) \rangle$  is zero and second cumulant is

$$\langle f(t) f(t') \rangle = \frac{1}{\tau_c} \delta(t - t') \tag{C.2}$$

[the factor  $\tau_c^{-1}$  in (C.2) was chosen only for later convenience]. We can explicitly solve (C.1), and using its solution and (C.2), we obtain

$$\langle \{X(t + \tau) - X(t)\}^2 \rangle = 1 - e^{-\tau/\tau_c} \tag{C.3}$$

$$\left\langle \left\{ \frac{1}{\tau} \int_t^{t+\tau} dt' X(t') \right\}^2 \right\rangle = \left( \frac{\tau_c}{\tau} \right)^2 \left( e^{-\tau/\tau_c} - 1 + \frac{\tau}{\tau_c} \right) \tag{C.4}$$

$$\left\langle \left\{ \frac{1}{\tau} \int_t^{t+\tau} dt' f(t') \right\}^2 \right\rangle = \frac{1}{\tau \tau_c} \tag{C.5}$$

On the other hand, the integration of (C.1) over  $(t, t + \tau)$  yields

$$X(t + \tau) - X(t) = -\frac{\tau}{\tau_c} \left\{ \frac{1}{\tau} \int_t^{t+\tau} dt' X(t') \right\} + \tau \left\{ \frac{1}{\tau} \int_t^{t+\tau} dt' f(t') \right\} \quad (C.6)$$

From (C.3)–(C.5) we see that (1) if  $\tau \ll \tau_c$ , the second term on the rhs is  $\sim (\tau/\tau_c)^{1/2} \ll 1$  and balances with the lhs, while (2) if  $\tau \gg \tau_c$ , the former is  $\sim (\tau/\tau_c)^{1/2} \gg 1$  and almost completely cancels with the first term on the rhs.

**APPENDIX D**

We evaluate (3.26) in the text for the concrete model of  $\mathcal{J}(z)$  and  $M(x)$ , which are defined as

$$\mathcal{J}(z) = \mathcal{J}_0 \exp \left( -\frac{\rho_0}{\rho_{eq} - z} + \frac{\rho_0}{\rho_{eq}} \right) \quad (D.1)$$

$$M(x) = \frac{1}{2l_M} \exp \frac{-|x|}{l_M} \quad (D.2)$$

where  $\mathcal{J}_0$ ,  $\rho_0$ , and  $\rho_{eq}$  are positive constants. From the discussion leading to (3.29), the lower bound of integration in (3.36) can be set equal to zero. Given the forms of  $\mathcal{J}(z)$  and  $M(x)$  as above, (3.36) can be rewritten, apart from the cutoff factor  $\theta(l_M^{-1} - |k|)$ , as follows

$$S_k(t) = \rho_{eq} R(t) \int_0^1 dy \exp \left( \frac{2b\tilde{M}_k}{1 - R(t)y} - \frac{2b\tilde{M}_k}{1 - R(t)} \right) \quad (D.3)$$

where

$$R(t) = \rho^*(t)/\rho_{eq} \quad (D.4)$$

$$b = \rho_0/\rho_{eq} \quad (D.5)$$

$$\tilde{M}_k = 1/(1 + k^2 l_M^2) \quad (D.6)$$

We can evaluate the integral in the two limiting cases  $2b\tilde{M}_k \gg 1$  (case A) and  $2b\tilde{M}_k \ll 1$  (case B):

$$S_k(t) \approx \begin{cases} \rho_{eq} R(t) & \\ \text{for } \begin{cases} R(t) \lesssim \frac{1}{2b\tilde{M}_k} & \text{(case A)} \\ 1 - R(t) \gg 2b\tilde{M}_k & \text{(case B)} \end{cases} & \\ \frac{\rho_{eq} [1 - R(t)]^2}{2b\tilde{M}_k} & \\ \text{for } \begin{cases} \frac{1}{2b\tilde{M}_k} \lesssim R(t) \leq 1 & \text{(case A)} \\ 1 - R(t) \ll 2b\tilde{M}_k \left\{ \log \frac{2b\tilde{M}_k}{[1 - R(t)]^2} \right\}^{-1} & \text{(case B)} \end{cases} & \end{cases} \quad (D.7)$$

In both cases we can construct the following approximant for  $S_k(t)$ :

$$S_k(t) \simeq \rho_{\text{eq}} \left\{ \frac{1}{R(t)} + \frac{2b\tilde{M}_k}{[1-R(t)]^2} \right\}^{-1} \quad (\text{D.8})$$

Substituting (D.6) into (D.8), we obtain

$$S_k(t) \simeq \rho^*(t) \left\{ 1 - \frac{\phi(t)}{1+\phi(t)} \frac{1}{1+k^2 l_{\text{eff}}(t)^2} \right\} \quad (\text{D.9})$$

where

$$\phi(t) = \frac{2bR(t)}{[1-R(t)]^2} = \frac{2\rho_0\rho^*(t)}{[\rho_{\text{eq}} - \rho^*(t)]^2} \quad (\text{D.10})$$

$$l_{\text{eff}}(t) = \frac{l_M}{[1+\phi(t)]^{1/2}} \quad (\text{D.11})$$

The term containing  $\phi(t)$  in (D.9) describes the correlation in the particle density. The density correlation function is given by

$$\langle \delta\rho(x, t) \delta\rho(x+r, t) \rangle \equiv \rho^*(t) [\delta(r) + w(r, t)] \quad (\text{D.12})$$

with

$$w(r, t) = \int \frac{dk}{2\pi} [S_k(t) - \rho^*(t)] \exp(ikr) \quad (\text{D.13})$$

$$= -\frac{\phi(t)}{1+\phi(t)} \frac{1}{2l_{\text{eff}}(t)} \exp \frac{-|r|}{l_{\text{eff}}(t)} \quad (\text{D.14})$$

We see that a correlation hole occurs with size  $\sim l_{\text{eff}}(t)$  ( $\leq l_M$ ). Since  $\phi(t)$  increases with  $\rho^*(t)$ , the hole becomes narrower and deeper as time goes on. The existence of the correlation hole is the result of the negative feedback mechanism in the  $\rho^*(t)$ -dependent nucleation rate.

## APPENDIX E

When we can choose the coarse-graining scale  $l$  and  $\tau$  such that (3.12), (3.13), and (3.22) are satisfied and the  $l \lesssim l_M$ , we can derive the evolution of the coarse-grained density  $\rho(x, t)$  by the following path-integral formalism.

We define the unit cell  $\Delta_{xT}$  by

$$\Delta_{xT} = \{(x, t); (X-1/2)l \leq x \leq (X+1/2)l, T\tau < t \leq (T+1)\tau\} \quad (\text{E.1})$$



where  $X$  and  $T$  are integers. Under the above assumptions the nucleation rate within each unit cell can be regarded as constant and therefore the probability distribution  $P(N_{XT}; \rho)$  for the number of nucleation  $N_{XT}$  within the unit cell  $A_{XT}$  obeys the Poisson distribution,

$$P(N_{XT}; \rho) = \exp(-\bar{N}_{XT}) \frac{\bar{N}_{XT}^{N_{XT}}}{N_{XT}!} \tag{E.2}$$

$$\bar{N}_{XT} = \mathcal{I}(M * \rho(Xl, T\tau)) \quad l\tau \gg 1 \tag{E.3}$$

where  $\rho(x, t)$  is the coarse-grained density defined in the same manner as  $\delta\rho(x, t)$  [see (3.10)] and the last inequality is just the assumption (3.22). We note that  $\rho(Xl, T\tau)$  is determined by the previous nucleation events as follows:

$$\rho(Xl, T\tau) = l^{-1} \sum_{T'=0}^{T-1} N_{XT'} \tag{E.4}$$

Therefore the joint probability  $P\{N_{XT}\}$  that there are  $N_{XT}$  of the nucleation events in each unit cell  $A_{XT}$  is given by

$$P\{N_{XT}\} = \prod_T \prod_X P(N_{XT}; \rho) \tag{E.5}$$

with  $P(N_{XT}; \rho)$  depending not only on  $N_{XT}$ , but also on  $\{N_{X'T'}: T' < T\}$ . Using Stirling's formula for  $N_{XT}$ , we have

$$-\log P\{N_{XT}\} = l\tau \sum_{X'} \sum_{T'} \left\{ \frac{\bar{N}_{X'T'}}{l\tau} - \frac{N_{X'T'}}{l\tau} \left[ \log \left( \frac{\bar{N}_{X'T'}}{N_{X'T'}} \right) + 1 \right] \right\} \tag{E.6}$$

We now return to the continuum representation. We denote

$$l\tau \sum_X \sum_T \rightarrow \int dx \int dt \tag{E.7}$$

$$\frac{\bar{N}_{XT}}{l\tau} = \mathcal{I}(M * \rho(Xl, T\tau)) \rightarrow \mathcal{I}(M * \rho(x, t)) \tag{E.8}$$

$$\frac{N_{XT}}{l\tau} \rightarrow s(x, t) \tag{E.9}$$

where  $\rho(x, t)$  and  $s(x, t)$  are related via the coarse-grained form of (3.1), that is,

$$\frac{\partial}{\partial t} \rho(x, t) = s(x, t) \tag{E.10}$$

Then (E.6) is converted into

$$P\{N_{XT}\} \rightarrow P[s] = \exp\{-\Phi[s]\} \quad (\text{E.11})$$

$$\Phi[s] = \int dt \int dx \left\{ \mathcal{I}(M * \rho(x, t)) - s(x, t) \left[ \log \left( \frac{\mathcal{I}(M * \rho(x, t))}{s(x, t)} \right) + 1 \right] \right\} \quad (\text{E.12})$$

Equations (E.10)–(E.12) describe the path probability for the macroscopic nucleation source  $s(x, t)$ .

We are interested in the path probability functional  $\mathcal{P}[\rho]$  for the density  $\rho(x, t)$ . In our case  $\mathcal{P}[\rho]$  is proportional to  $P[s]$ , since  $s$  and  $\rho$  are linearly related by (E.10) and therefore the functional Jacobian (or the Radon–Nikodym derivative)  $D[\rho]/D[s]$  is independent of  $s$  or  $\rho$ . Thus we have

$$\mathcal{P}[\rho] = \exp\{-\varphi[\rho]\} \quad (\text{E.13})$$

$$\varphi[\rho] = \Phi[s] + \text{const} \quad (\text{E.14})$$

Since  $N_{XT} \gg 1$  [see (E.3)], the Poisson distribution  $P(N_{XT}; \rho)$  has a sharp maximum at  $N_{XT} = \bar{N}_{XT}$ . Then we can approximate  $\varphi[\rho]$  by its minimum plus the harmonic expansion around it, that is,

$$\varphi[\rho] \simeq \varphi[\rho^*] + \frac{1}{2} \int d(1) \int d(2) \frac{\delta^2 \varphi}{\delta \rho(1) \delta \rho(2)} \Big|_{\rho=\rho^*} [\rho(1) - \rho^*(1)][\rho(2) - \rho^*(2)] \quad (\text{E.15})$$

where  $\rho^*(x, t)$  is the minimum of  $\varphi[\rho]$  (the most probable path) and  $(i)$  with  $i=1$  or  $2$  denotes  $(x_i, t_i)$ . We determine  $\rho^*(x, t)$  from

$$\frac{\delta \varphi}{\delta \rho(x, t)} \Big|_{\rho=\rho^*} = 0 \quad (\text{E.16})$$

The lhs of (E.16) can be calculated using the following identity:

$$\frac{\delta}{\delta \rho(x, t)} = - \frac{\partial}{\partial t} \frac{\delta}{\delta s(x, t)} \quad (\text{E.17})$$

The result is

$$\frac{\delta \varphi}{\delta \rho(x, t)} = \frac{\partial}{\partial t} \log \left( \frac{\mathcal{I}(M * \rho(x, t))}{s(x, t)} \right) + M * \left\{ \mathcal{I}'(M * \rho) \left[ 1 - \frac{\mathcal{I}(M * \rho)}{s} \right] \right\} (x, t) \quad (\text{E.18})$$

where the last term on the rhs means that

$$M * \mathcal{O}(x, t) \equiv \int M(y-x) \mathcal{O}(y, t) dy \tag{E.19}$$

$$\mathcal{O}(y, t) \equiv \mathcal{I}'(M * \rho(y, t)) \left[ 1 - \frac{\mathcal{I}(M * \rho(y, t))}{s(y, t)} \right] \tag{E.20}$$

From (E.10), (E.16), and (E.18)–(E.20) the most probable path is given by

$$\frac{\partial}{\partial t} \rho^*(x, t) = \mathcal{I}(M * \rho^*(x, t)) \tag{E.21}$$

The path probability for the small fluctuation

$$\delta\rho(x, t) = \rho(x, t) - \rho^*(x, t) \tag{E.22}$$

is obtained from the second term in (E.15). After some simple algebra we obtain

$$\begin{aligned} & \left. \frac{\delta^2 \varphi}{\delta\rho(x, t) \delta\rho(x', t')} \right|_{\rho=\rho^*} \\ &= \frac{\partial^2}{\partial t \partial t'} \left\{ \frac{\delta(x-x')}{\mathcal{I}(M * \rho^*(x, t))} \delta(t-t') \right\} \\ &+ \frac{\partial}{\partial t} \left\{ \frac{\mathcal{I}'(M * \rho^*(x, t))}{\mathcal{I}(M * \rho^*(x, t))} M(x-x') \delta(t-t') \right\} \\ &+ \frac{\partial}{\partial t} \left\{ \frac{\mathcal{I}'(M * \rho^*(x, t))}{\mathcal{I}(M * \rho^*(x, t))} M(x'-x) \delta(t-t') \right\} \\ &+ \int dy \frac{[\mathcal{I}'(M * \rho^*(y, t))]^2}{\mathcal{I}(M * \rho^*(y, t))} M(y-x) M(x'-y) \delta(t-t') \end{aligned} \tag{E.23}$$

Substituting this into (E.15) and integrating by parts, we obtain, apart from the boundary terms,

$$\begin{aligned} \varphi[\rho] \simeq & \varphi[\rho^*] + \int dx \int dt \frac{1}{2\mathcal{I}(M * \rho^*(x, t))} \\ & \times \left\{ \frac{\partial}{\partial t} \delta\rho(x, t) - \mathcal{I}'(M * \rho^*(x, t)) M * \delta\rho(x, t) \right\}^2 \end{aligned} \tag{E.24}$$

Expressions (E.13) and (E.24) mean that in the present approximation

$$f(x, t) \equiv \frac{\partial}{\partial t} \delta\rho(x, t) - \mathcal{I}'(M * \rho^*(x, t)) M * \delta\rho(x, t) \tag{E.25}$$

is the Gaussian white noise with second cumulant

$$\langle f(x, t) f(x', t') \rangle_c = \mathcal{I}(M * \rho^*(x, t)) \delta(x - x') \delta(t - t') \quad (\text{E.26})$$

Equations (E.25) and (E.26) recover the result for the intermediate stage obtained in Section 3.

## APPENDIX F

We argue that  $F_b(n, t)$  given in (4.21) is an attractor of the solution of (4.19) and (4.20). Defining  $\phi(n, \log t) = F(n, t) - F_b(n, t)$ , the linearized evolution equation for  $\phi(n, \lambda)$  becomes

$$\frac{\partial}{\partial \lambda} \phi(n, \lambda) = \left\{ \frac{n(N-n)}{2} \frac{\partial^2}{\partial n^2} - 1 \right\} \phi(n, \lambda), \quad 0 \leq n \leq N \quad (\text{F.1})$$

with the boundary conditions

$$\phi(0, \lambda) = \phi(N, \lambda) = 0 \quad (\text{F.2})$$

We consider the quantum mechanical Hamiltonian

$$\hat{H} \equiv \frac{1}{2} \hat{p}_n^2 + (E+1)/n(N-n), \quad 0 \leq n \leq N \quad (\text{F.3})$$

where  $\hat{p} = (-i) \partial/\partial n$  and  $E$  is real. It corresponds to the assumption that in (F.1) there is no oscillatory varying modes, which seems natural for the present relaxation process. Then we see clearly that for  $E+1 > 0$  all the eigenvalues of  $\hat{H}$  must be positive. This means that (F.1) and (F.2) cannot have a solution of the type  $\phi(n, \lambda) = \psi(n) \exp(E\lambda)$  with  $E > -1$ .

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## REFERENCES

1. K. Tsuda, N. Yamamoto, K. Yagli, and K. Hamano, in *International Congress on Electron Microscopy, Kyoto* (1986), Vol. II, p. 1233.
2. M. Glogarova, L. Lejcek, J. Pavel, V. Janovec, and J. Fousek, *Mol. Cryst. Liq. Cryst.* **91**:309 (1983); H. Orihara and Y. Ishibashi, *Ferroelectrics* **58**:179 (1984).
3. K. Kawasaki, K. Sekimoto, and S. Yamanaka, *Physica* **23D**:122 (1986).

4. R. L. Stratonovich, *Topics in the Theory of Random Noise* (Gordon and Breach, 1983), Vol. 1.
5. R. Kubo, K. Matsuo, and K. Kitahara, *J. Stat. Phys.* **9**:51 (1973).
6. H. Mori and K. J. McNeil, *Prog. Theor. Phys.* **57**:770 (1977).
7. T. Morita, H. Mori, and M. Tokuyama, *J. Stat. Phys.* **18**:137 (1978).
8. E. D. Siggia and A. Zippelius, *Phys. Rev. A* **24**:1036 (1981); see also K. Kawasaki, *Physica* **124B**:156 (1984).
9. K. Sekimoto, *Prog. Theor. Phys.* **71**:885 (1984).