Stochastic Process of Nucleation in the Early Stage

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Stochastic theory of self-organization in the early stage is studied based on the birth-and-death type master equation. Universal relations are derived concerning the time variation of fluctuations. An adsorption-desorption process, which has a quadratic nonlinearity, is studied with computer experiments and the system size dependence of fluctuation is obtained numerically.

§ 1. Introduction

Self-organization is one of the most remarkable phenomena in the far from thermal equilibrium state of open systems. Those macroscopic orders in dissipative systems are understood as the breakdown of the temporal and/or spatial symmetry. The occurrence of the ordered states is induced by the destabilization of the primary thermodynamical branch and the stabilization of a bifurcating branch. As instability of the primary branch is accompanied by increase of the fluctuation in the system, the deterministic path cannot be clearly decoupled from the time course of fluctuations any longer. That is to say, the physical meanings of the deterministic description become vague near bifurcation points and the stochastic description must be used. This situation is considered a kind of chaos and is called "generalized turbulence." New ordered state is organized through this chaotic regime. The purpose of this article is to study this regime.

The early stage process of self-organization through fluctuation should be analyzed by stochastic approaches. The relaxation processes from unstable or metastable states are simple examples of self-organization through fluctuation. In this article discussion is limited to the Markoffian processes described by the following birth-and-death master equation,

\[ \frac{\partial P}{\partial t} = - \sum_{R}(W(X, R)P(X, t) - W(X - R, R)P(X - R, t)), \]

(1)

where \( X \) is a macrovariable, \( P(X, t) \) and \( W(X, R) \) are probability density function (p.d.f.) and transition probability with step \( R \), respectively. As is known well, Eq. (1) is rewritten into the Kramers-Moyal expansion,

\[ \frac{\partial p}{\partial t} = \sum_{n} \frac{1}{n!} \left( - \frac{\partial}{\partial x} \right)^n \psi_n(x) p(x, t), \]

(2)
where the following relations are used, \( p = PN, \quad x = X/N \) and \( c_n = \sum_s R^s W(Nx, R)/N^s \). Here \( c_n(X) \) is of \((n-1)\)-th order of inverse system size \( N^{-1} \). Let’s consider the potential function \( \phi(x, t) = -\log (p(x, t)) \). Figure 1 is the conceptual simple bifurcation diagram which shows the time course of \( \phi(x, t) \) from the initial time \( (t = t_0) \) to infinitely far future \( (t = t_\infty) \). Initial unstable p.d.f. \( p(x, t_0) \) relaxes to a stable steady p.d.f. \( p(x, t_\infty) \). In the initial and the final stage \( (t < t_\beta \) and \( t > t_\beta) \), \( \phi(x, t) \) has a minimum and in the intermediate stage \( (t_\beta < t < t_\delta) \), \( \phi(x, t) \) has double minima. The most probable states corresponding to each phase change along the lines IB and AF. At the time when the double minima of \( \phi(x, t) \) get equal depth, macroscopic phase separation may be observed. In a stochastic sense, however, the second phase comes to appear in the system suddenly at A-point, and the 1st phase disappears at B-point. In the present paper A-point \( (x_\alpha, t_\alpha) \) is called nucleation point in a stochastic sense, and we will limit our analysis to the fluctuation around this dip of potential \( \phi(x, t) \). The appearance of the nucleation point as mentioned above is general for the case of relaxation from a metastable state. But in the case of relaxation from unstable state the appearance of two most probable peaks in p.d.f. is not so general. Namely, in some cases the most probable path may be single for all time, and in the case when the points A and B in Fig. 1 degenerate the critical fluctuation appears in the system. In this paper we will mainly concern to the relaxation process from an unstable state. In this process there seems to be two characteristic times; the one is the characteristic time \( T_1 \) of the fading process of initial most probable peak, which becomes longer when the initial state approaches to the unstable state, and the other is the characteristic time \( T_2 \) of the self-organization process of the second most probable peak, which is determined mainly by the proper characters of the system under consideration. When \( T_1 \ll T_2 \), p.d.f. has a well-defined sharp peak, a systematic method is applicable. When \( T_1 \approx T_2 \), however, the systematic method fails completely as reported by Suzuki. Furthermore, in the

Fig. 1. Conceptual diagram of a simple bifurcation.
case $T_1 > T_2$ the phase exchange may appear as shown in Fig. 1, which we are going to study in the present paper. As easily noticed, the systematic size expansion is applicable usefully only in the restricted regime of $T_1/T_2 \ll 1$ and $T_1/T_2 \gg 1$. Near the nucleation points as A and B explained in Fig. 1, however, the system size dependence of fluctuation is delicate problem.

In § 2 dynamical equations of nucleation are derived, and is solved in § 3. Section 4 is devoted to the numerical study of the simple example of the relaxation process from unstable state, where the size dependence of nucleation process is discussed. Section 5 is devoted to discussion.

§ 2. Dynamical equations of nucleation

The master equation (2) is rewritten into the following formal expansion,

$$
\frac{-\partial \phi}{\partial t} = \left( -\frac{\partial c_1}{\partial x} + \frac{1}{2!}\frac{\partial^2 c_2}{\partial x^2} - \frac{1}{3!}\frac{\partial^3 c_3}{\partial x^3} + \cdots \right) + \left( -c_1 + \frac{\partial c_1}{\partial x} - \frac{1}{2!}\frac{\partial^2 c_2}{\partial x^2} + \cdots \right) \left\{ -\frac{\partial \phi}{\partial x} \right\}
$$

$$
+ \left( \frac{1}{2!} c_2 - \frac{1}{2!}\frac{\partial c_2}{\partial x} + \cdots \right) \left\{ \left( \frac{\partial \phi}{\partial x} \right)^2 - \frac{\partial^2 \phi}{\partial x^2} \right\}
$$

$$
+ \left( -\frac{1}{3!} c_3 + \cdots \right) \left\{ -\left( \frac{\partial \phi}{\partial x} \right)^3 + 3\left( \frac{\partial \phi}{\partial x} \right) \left( \frac{\partial^2 \phi}{\partial x^2} \right) - \frac{\partial^3 \phi}{\partial x^3} \right\}
$$

$$
+ \cdots . \quad (3)
$$

As was already mentioned, the most dominant contribution in the limit of $N \to \infty$ comes from the first term in each bracket ( ). We will take into account only the dominant terms at thermodynamic limit because the main concern of this paper is to get the asymptotic relation in the limit of large $N$. It is important, however, that the extensive property of $\phi \sim O(N)$ is not assumed a priori in the present article. In other words, the potential $\phi$ satisfies the following relation,

$$
\frac{-\partial \phi}{\partial t} = -\frac{\partial c_1}{\partial x} + \sum_{n=1}^{\infty} (-1)^n \frac{c_n}{n!} \left\{ \left( \frac{\partial}{\partial x} \right)^n e^{-\phi} \right\}. \quad (4)
$$

At the peak point of p.d.f., $x = y(t)$, $\phi$ is expressed by the power series expansion,

$$
\phi(x, t) = \sum_{k=0}^{\infty} a_k(t) (x - y(t))^k. \quad (5)
$$

Here $a_k(t)$ is the expansion coefficient, and $a_1(t) = 0$ is always satisfied since $x = y(t)$ is a minimum point of $\phi(x, t)$. By putting Eq. (5) into Eq. (4), and
equating each term including the same power of \((x - y(t))^k\) on both sides, we can obtain a series of the coupled differential equations of \(a_k(t)\) and \(y(t)\). Then we have to expand \(c_n(x)\) and \(\frac{\partial c_n(x)}{\partial x}\) into power series of \((x - y(t))\), for example,

\[
c_n(x) = \sum_{k=0}^{\infty} \frac{1}{k!} \frac{\partial^k c_n(y)}{\partial y^k} (x - y(t))^k, \quad \text{etc.} \quad (6)
\]

In what follows, we will use the following notation,

\[
\frac{\partial^k c_n}{\partial y^k} = \frac{\partial^k c_n(x)}{\partial x^k} \bigg|_{x=y}.
\]

Near the peak point \(x = y(t)\) the dynamics of fluctuations is described by the time courses of \(a_k(t)\) and \(y(t)\),

\[
\begin{align*}
-\frac{da_k}{dt} &= \mathcal{F}_0(y, \{a_j\}), \\
2a_k \frac{dy}{dt} &= \mathcal{F}_1(y, \{a_j\}), \\
(k+1)a_{k+1} \frac{dy}{dt} &= \frac{da_k}{dt} + \mathcal{F}_k(y, \{a_j\}),
\end{align*}
\]

(7)

where \(\mathcal{F}_k(y, \{a_j\})\) stands for the expansion coefficient of the power series of the r.h.s. in Eq. (4), i.e., for example,

\[
\mathcal{F}_1(y, \{a_j\}) = -\frac{\partial^2 c_1}{\partial y^2} + 2c_1a_s + \left(3\frac{\partial c_2}{\partial y} - 3c_2\right)a_s + \frac{\partial c_3}{\partial y}a_s + \cdots. \quad (8)
\]

As one can see easily, \(\partial c_n/\partial y \ll c_{n-1}\) (for large \(N\)), therefore, the dominant contribution of \(dy/dt\) is rewritten as follows,

\[
\frac{dy}{dt} = \{c_1 - a_2c_2 + 4a_4c_4 - O(c_6)\}
\]

\[
-\frac{1}{2a_s} \left[ \frac{\partial^2 c_1}{\partial y^2} + 3a_2c_2 - 4a_4c_4 + 5a_6c_6 + O(c_8) \right]. \quad (9)
\]

As mentioned in the previous section the value of \(a_2(t)\) becomes zero at the nucleating point \((t_0, x_0)\). Clearly then, the second term contributes dominantly in the early stage. That is to say,

\[
\frac{dy}{dt} = \frac{1}{2a_s} \left[ \frac{\partial^2 c_1}{\partial y^2} + 3a_2c_2 - 4a_4c_4 + 5a_6c_6 + O(c_8) \right] = \frac{G(y, \{a_j\})}{2a_s}. \quad (10)
\]

Here \(O(c_6)\) gives the contribution coming from \(c_k\)'s \((k \geq 5)\). By the same consideration used in the derivation of Eq. (10), the equation of \(a_k\) is rewritten
into the following form,
\[
\frac{da_k}{dt} = (k+1) a_{k+1} \frac{dy}{dt}.
\]
(11)

Namely, the dynamics of the early stage fluctuations obeys the following universal law,
\[
\frac{da_k}{dt} = \frac{(k+1) a_{k+1} a_{k+1}}{(k+2) a_{k+2}} \frac{dy}{dt},
\]
\[
\frac{dy}{dt} = a_2 \cdot \frac{da_2}{dt}.
\]
(12)

This formula is free from the characteristics of the system under consideration.

§ 3. The time course of the fluctuation in the early stage

The solutions of \(a_2\) and \(y\) is obtained from Eqs. (10) and (11),
\[
a_2(t) = \sqrt{K(t-t_c)},
\]
\[
y(t) = x_c + \frac{1}{3 a_2^3} \sqrt{K(t-t_c)},
\]
(13)

where
\[
K = 3 a_2^3 G(x_c, \{a_j^0\}) \text{ and } a_j^0 = a_j(t_c).
\]

The birth or death of the nucleus in the distribution function is described in the case of \(K>0\) or \(K<0\), respectively. As noticed easily when \(a_k^0\) happens to be zero, the solution diverges to infinity. Therefore, we must look for another physically meaningful solution. To this end we will consider the following general case: \(a_k^0(k=2, 3, \ldots, n+1)\) are zero but \(a_{n+2}^0\) is non-zero and finite. Then the solution of Eq. (12) is given by
\[
a_k(t) = A_k(t-t_c)^{\mu_k}, \quad y(t) = x_c = \hat{H}(t-t_c)^{\nu},
\]
\[
\nu = 1/(n+1), \quad \mu_k = (n-k+2)/(n+1).
\]
(14)

Here,\[
\hat{H} = -(n+1) G(x_c, \{a_j^0\}) / \alpha,
\]
\[
A_k = \alpha \left( -\frac{\alpha}{G(x_c, \{a_j^0\})} \right)^{k-2} (\mu - 1/2) (\mu - 2/3) \cdots (\mu - k-3/2) / k(k-1),
\]

where the following use were made of \(2A_k = \alpha, \mu_k = \mu\). \(A_k\) and \(\hat{H}\) stand for the growth rates of new phase. Generally speaking, these quantities are not
always of order $N$ in the early stage. In the latter section computer experiments will give some predictions about the system size dependence of these quantities. When $a_{k'}(k' = n + 2, n + 3, \cdots, m)$ are non-zero and finite (here we assume $a_{m+1} = 0$ implicitly), the solutions of $a_{k'}$ are obtained by the same technique used in the derivation of Eq. (14),

$$a_{k'}(t) = a_{k'} + A_{k'}(t - t_e)^{n_{k'}},$$

$$\mu_{k'} = \mu_{n+1} = 1/(n + 1),$$

$$a_{k'+1} = \frac{n + 2}{k' + 1} \frac{A_{k'}}{A_{n+1}}. \ (k' = n + 1, \cdots, m - 1) \quad (15)$$

These are not the complete solution of $a_{k'}$, since we can know only the ratio $a_{k'}/A_{k'-1}$, and $\mu_m$ is not defined. In order to get the complete solutions, we have to analyse Eqs. (12) by the higher order approximation. The higher order perturbation analysis is beyond the purpose of the present article.

At the last part of this section we will discuss the effect arising from the truncation of the potential $\phi$. Let’s consider the case where $\phi$ is approximated by a polynomial,

$$\phi(x, t) = \sum_{k=0}^{i} a_k(t) (x - y(t))^k. \quad (16)$$

By this truncation of $\phi$ the results obtained so far is modified only a little. Since $a_{i+1} = 0$, the solution of $a_i$ is given from Eq. (7) as follows,

$$a_i(t) = a_i(t) + A_i(t - t_e). \quad (17)$$

Therefore the index $\mu_i$ is unity.

\section*{§ 4. Computer experiments}

Let’s consider the adsorption-desorption process on a membrane which is soaked in a solution. On the membrane there are distributed $N$ active sites that adsorb the molecule of a chemical substance $C$ solving in the solution. For the sake of simplicity we assume that each site can adsorb only one $C$-molecule. Let’s analyse the adsorption-desorption process described by the following rate equation,

$$\frac{dx}{dt} = k_1cx(1 - x) - k_2x + k_3c(1 - x), \quad (18)$$

where $c$ and $x$ denote the concentration of the chemical substance $C$ and the fraction of the site occupied by the molecule. $k_i$ is rate constants. The stochastic model of this rate process is constructed by giving the transition probability $W(X, R)$ as follows,
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\[ W(X, 1) = S_1 C X (N - X) + S_2 C (N - X), \]
\[ W(X, -1) = S_3 X, \]  
where \( S_i \) is the corresponding constant, and the master equation is written by

\[
\frac{\partial P}{\partial t} = \{W(X + 1, -1) P(X + 1, t) + W(X - 1, 1) P(X - 1, t)
- \{W(X, -1) P(X, t) + W(X, 1) P(X, t)\}. \]  

(20)

Here \( P(X, t) \) is the p.d.f., \( C \) and \( X \) denote the total number of the molecule \( C \) and of the site occupied by the \( C \)-molecule. \( C, X \) and \( W(X, R) \) are the quantities of order \( N^1 \). Clearly then the generalized Fokker-Planck equation is derived as follows,

\[
N^{-1} \frac{\partial p}{\partial t} = k_1 c \left( x - \frac{1}{N} \right) \left( 1 - x + \frac{1}{N} \right) p \left( x - \frac{1}{N} \right)
- k_2 x p(x) + k_3 c \left( 1 - x + \frac{1}{N} \right) p \left( x - \frac{1}{N} \right)
- \left[ k_1 c x (1 - x) p(x) - k_2 \left( x + \frac{1}{N} \right) p \left( x + \frac{1}{N} \right) + k_3 c (1 - x) p(x) \right],
\]  

(21)

where \( p(x) \) is the p.d.f. about the concentration \( x \) and the following scalings were used,

\[
x = X/N, \quad c = C/N, \quad p(x) = NP(Nx, t),
S_1 = k_1/N^2, \quad S_2 = k_2, \quad S_3 = k_3/N.
\]  

(22)

The steady state solution of \( P(X, \infty) \) is given by

\[
P(X, \infty) = P(0) \left( \frac{S_1 C}{S_3} \right)^x \left( \frac{N}{X} \right)^x \Gamma \left( X + \frac{S_2}{S_1} \right).
\]  

(23)

Here \( P(0) \) is a normalization parameter. When \( S_3 = 0 \), the unique steady solution of \( P(X, \infty) \) is \( \delta_{X,0} \). This steady distribution cannot reproduce the

![Fig. 2. Time courses of \( x, \langle x \rangle \) and \( y \).](image-url)
macroscopic stable solution derived from the rate equation. This situation is known as the “absorbing boundary phenomena”. In the present paper we limit our discussion to the time dependent phenomena in the case of $1 \gg k_0 > 0$. Let’s start with the result of computer simulation. The time course of the relaxation process of this model is shown in Fig. 2. $x(t)$ denotes the solution of the rate equation (18) under the initial condition of $x(0) = 0$, $\langle x(t) \rangle$ is the mean value calculated by

$$\langle x(t) \rangle = \int_0^1 x p(x) dx$$

and $y(t)$ is the coordinate of the small peak point newly appeared in the p.d.f. $p(x, t)$ as mentioned in § 1. Here the initial distribution $P(X, 0)$ is settled in $P(X, 0) = \delta_{x_0}$ corresponding to the macroscopic situation of $x(0) = 0$. The following parameters are used,

$$k_1 = k_2 = 1, \quad k_3 = 10^{-4},$$

$$C = 2N, \quad N = 100.$$  

When the value of $S_0$ goes to small, the difference between the deterministic path $x(t)$ and mean value $\langle x(t) \rangle$ increases. This situation has already been indicated as “anomalous relaxation from an unstable state”. In our calculation, on the way of the relaxation of the initial unstable peak, the another small peak come to appear and it grows gradually. Namely in the intermediate stage there exist two peaks in the p.d.f. The initial peak diminishes monotonically and disappears finally, on the other hand the second small peak increases and takes the place of the initial dominant peak. The point is that the second

Fig. 3. Time courses of fluctuation. Each scale is plotted in arbitrary unit.
small peak comes to appear suddenly in the system at the onset point \((t_0, x_0)\).

The precise behavior of the fluctuation around the onset point of the second peak is shown in Fig. 3. The time courses of each variable are plotted in arbitrary unit scale. Here the potential is approximated by the 5-th order polynomial;

\[
\phi(x, t) = \sum_{k=0}^{5} a_k(t) (x - y(t))^k.
\]  

(25)

The dotted lines in Fig. 3 stand for the value of each coefficient at the inflection point of \(\phi(x, t)\) as mentioned in §1. By the method of curve fitting we can determine the values of time indices \(\mu, \nu\) and growth rate \(A_k\) as follows,

Fig. 4. System size dependence of \(A_k\).

Fig. 5. System size dependence of \(a_k\).

Fig. 6. System size dependence of nucleation point.
\[ \nu = \mu_e = \mu_e = 1/2, \quad \mu_e = 1. \]  

(26)

These are well line with the theoretical results mentioned in § 3.

At the last of this section we will discuss the system size dependence of the fluctuation in the early stage. If we change the value of the system size \( N \), time indices obtained in Eq. (26) are almost invariant. On the other hand the values of other variables \( A_5 \) and \( a_4 \) depend on the system size sensitively but monotonically. Figures 4~6 show the results of the system size dependence of \( A_5 \), \( a_4 \) and nucleation point \( (t_\infty, x_\infty) \). Size dependence of \( A_5 \), \( A_4 \), \( a_4 \) and \( a_5 \) is monotonical and their dependence seems to be less than \( O(N^1) \), i.e.,

\[ A_5, A_4 \leq O(N^1) \quad \text{and} \quad a_4, a_5 \leq O(N^1). \]  

(27)

While the size dependence of \( A_5 \), \( A_4 \) and \( a_5 \) is not regularly monotonical. Presumably, the reason comes from the effect of truncation of potential as Eq. (25), and all these quantities may also be regularly monotonical function of \( N \) provided that we use higher order approximation of \( \phi(x, t) \). Figure 6 shows the size dependence of the onset point \( (t_\infty, x_\infty) \). In our calculation system size \( N \) is changed from 50 to 150. In this limited range the asymptotic value of \( t_\infty \) seems to increase as

\[ t_\infty \sim O(N^1) \]

and a minimum point of \( t_\infty \) appears around \( N = N_\infty = 60 \). The appearance of the minimum suggests that there exists a most effective size to develop a nucleus in the system. The problems about the effective size are beyond the purpose of the present paper based on the homogeneous fluctuation and will be discussed elsewhere taking account of the effect induced by diffusion.

§ 5. Discussion

In the present article we discussed the homogeneous fluctuation in the early stage of self-organization. Our theory proposed here seems to have a close connection with the theory of anomalous relaxation reported by Suzuki. It is, however, important to point out that the situation analysed in this article is essentially different from the anomalous enhancement of fluctuation. That is to say, our main concerns are not the analysis of the second order type bifurcation, but the analysis of the first order type phase transition. This is the reason why we called \( (t_\infty, x_\infty) \) nucleation point even though the analysis in § 4 is limited to the case of homogeneous relaxation from an unstable state. As easily noticed from Eq. (9), the motion of peak point in p.d.f. is described by

\[ \frac{dy}{dt} = f(y, \{a_j\}) - \frac{1}{2a_2} g(y, \{a_j\}). \]
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If the peak is sharp and well-defined, \( a_\varepsilon \gg 1 \) is satisfied and the contribution of the second term on the r.h.s. is negligible. Especially, when the state is in thermodynamical aged stage, i.e., \( a_\varepsilon \sim O(N) \), the deterministic path describing the peak point is decoupled from fluctuations, and is given by the aged stage evolution equation, \( \dot{\phi} = c_1(y) \).

At the nucleation point let's estimate the size dependence of partial variance around the second peak,

\[
\langle \Delta X^+ \rangle = \int (X - Y)^4 P(X, t_c) dX,
\]

where \( Y \) stands for the peak point, i.e., \( Y = N\varepsilon \). The dominant contribution comes from \( \phi(x, t_c) \approx a_\varepsilon^4 (X - Y)^4 \),

\[
\langle \Delta X^+ \rangle \sim O(N^4 / \sqrt{a_\varepsilon}) \geq O(N^4) .
\]

Here we used Eq. (27). This means the breakdown of central limit theorem in the early stage of self-organization.\(^9\) The theory developed here is applicable to the case of relaxation from a metastable state, then the onset time \( t_c \) is compared to the mean first passage time from the metastable state to a stable one.

References


Discussion

R. Kubo: If I understood correctly, you assume that there is a certain time \( t_c \) where you see a sign of growth at \( x=x_\varepsilon \) in the neighborhood of final equilibrium and start there by assuming \( \dot{\phi} = \sum a_\varepsilon (x - y(t))^4 \). How do you determine \( x_\varepsilon \)? Isn't it that you really start at \( t=t_c \) with an arbitrary assumption of the initial form of distribution function?

Y. Aizawa: Here I obtained universal dynamical equations describing the early stage fluctuation, but the initial condition of these equations is not determined uniquely, therefore, \( x_\varepsilon \), \( t_c \) and \( a_\varepsilon \) are parameters in my theory.
M. Suzuki: Can you determine the onset time $t_\alpha$ in your theory? If not, I think that you are starting effectively from the onset time as the initial time, as Prof. Kubo has just pointed out.

Y. Aizawa: Theoretically I could not. Results reported here in § 4 were obtained through computer calculation for complete master equation. From the theoretical viewpoint, we may solve the equation obtained here under arbitrary initial conditions at onset time $t_\alpha$, but as shown in Fig. 5, size dependence of $a_\alpha$ must be taken into account.

G. Nicolis: As I understood your paper, in the numerical simulation of the solution of the complete master equation, you find evidence of a second peak in the probability distribution. On the other hand, in the analytic treatment you have not yet taken into account the interaction between the initial peak around the unstable state and the secondary peak arising through nucleation. Is this the correct way of interpreting your results?

Y. Aizawa: Yes, you are right. My analysis is mainly limited to the case of $T_1 \gg T_2$, where we can generally expect the appearance of the second peak in the probability distribution. When $T_1$ and $T_2$ are in same order, as the result of the interference between the initial and secondary peaks, we can observe the anomalous enhancement of fluctuation as reported by Prof. Suzuki in this meeting.

M. Suzuki: I will give a comment in connection with the extensivity. The extensive part in the probability function exists always for a finite time, but it becomes smaller and smaller as time goes to infinity, when the system starts from the unstable point. Therefore, the remaining part becomes equally important to describe correctly the onset process of macroscopic order in my scaling theory of transient phenomena near the instability point.