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Yusuke Yanagisawa and Shin-ichi Sasa

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Phase coexistence in a weakly stochastic reaction-diffusion system

Yusuke Yanagisawa and Shin-ichi Sasa
Department of Physics, Kyoto University, Kyoto 606-8502, Japan
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We investigate phase coexistence in a weakly stochastic reaction-diffusion system without assuming a continuum description. Concretely, for $(2N+1)$ diffusion-coupled vessels in which a chemical reaction exhibiting bistability occurs, we derive a condition for the phase coexistence in the limit $N \to \infty$. We then find that the phase coexistence condition depends on the rate of hopping between neighboring vessels. The conditions in the high- and low-hopping-rate limits are expressed in terms of two different potentials which are determined from the chemical reaction model in a single vessel.

Introduction.— Macroscopic systems out of equilibrium exhibit various ordered structures called dissipative structures [1–4]. Although most of these studies are based on deterministic equations, fluctuation effects on microscopic and mesoscopic systems out of equilibrium have recently attracted much attention [5, 6] because advances in experimental techniques have made it possible to observe, manipulate and control microscopic and mesoscopic dynamics [7, 8]. Examples include fluctuation effects on Turing patterns [9–13], oscillation [14], chaos [15], phase coexistence [16], and front propagation [17–20]. In this Letter, we study fluctuation effects on phase coexistence in reaction-diffusion systems.

In particular, we focus on weakly stochastic systems. When there exists a unique stationary solution of the corresponding deterministic equation ignoring noise for the weakly stochastic system, the most probable value at the steady state is equal to the stationary solution. In contrast, when multiple stationary solutions exist for the deterministic equation, the most probable value would be determined as the stationary solution that maximizes the probability. This has been fully understood through the study of simple stochastic chemical reaction models [21, 22].

A similar but more complex situation occurs for reaction-diffusion systems. As an example, we consider a one-dimensional stochastic reaction-diffusion system where the chemical reaction has a bistable property. For this system, there is a special parameter value at which a spatially inhomogeneous stationary pattern appears. This pattern represents phase coexistence in one-dimensional space, as indicated by Schlögl [23]. More precisely, a stable homogeneous state corresponds to each phase, and a kink pattern connects the two stable states like an interface in the liquid-gas phase coexistence as shown in Fig. 1. When the system is described by a partial differential equation with weak noise, the condition for the phase coexistence is perturbatically obtained from that for the reaction-diffusion equation without noise [17, 18, 20]. However, when the interface width between the two phases is much smaller than the coarse-graining length of the continuum description, the validity of the continuum description is not ensured.

In this Letter, to study the phase coexistence condition for weakly stochastic reaction-diffusion systems without assuming a continuum description, we consider a stochastic process in $(2N+1)$ diffusion-coupled reaction vessels. We then find that the phase coexistence condition depends on the rate of hopping between the neighboring vessels. Remarkably, at the high-hopping-rate limit, the phase coexistence condition is obtained through the analysis of the reaction-diffusion equation, while at the low-hopping-rate limit, the phase coexistence condition is obtained through the perturbation analysis of the steady state. We show that these two conditions are expressed in terms of two different potentials, namely deterministic and stochastic potentials, which are determined from the chemical reaction model.

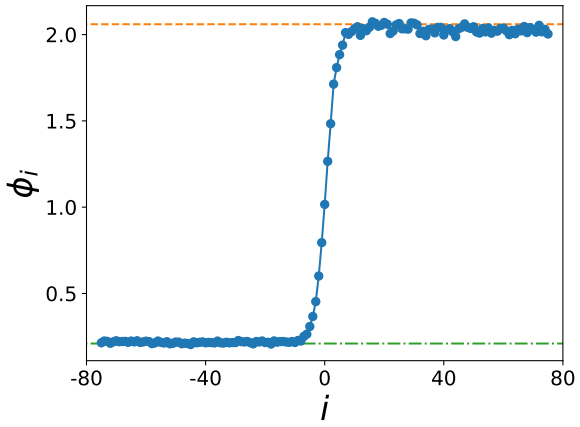


FIG. 1. Phase coexistence state of the stochastic reaction-diffusion system in the high-hopping-rate regime for $r = 5.0$. The orange dotted line and green dash-dot line indicate ξ_+ and ξ_-, respectively. See the paragraph including (14) for the explanation of the pattern.

Setup.— First, we study a well-mixed chemical reaction system in a reaction vessel. To be specific, we analyze the Schlögl model [23] described by

$$A \underset{k_{-1}}{\overset{k_{+1}}{\rightleftharpoons}} X, \tag{1}$$

$$3\,X \underset{k_{-2}}{\overset{k_{+2}}{\rightleftharpoons}} 2\,X + B. \tag{2}$$

Here, the concentrations of the chemical species A and B are fixed and we consider the dynamics of the concentration of the chemical species X. Let Ω be the volume

of the system. We use the same symbol X to denote the number of particles of the chemical species X. The concentrations of the chemical species A, B and X are denoted a, b and ξ, respectively. In the argument below, we set $a = 1$ and $k_{+1} = 0.5$ to make all quantities dimensionless.

We assume that the time evolution of X is described by the Markov jump process. The probability distribution $P_t(X)$ of X at time t obeys the master equation

$$\frac{\partial P_t(X)}{\partial t} = W_+(X-1)P_t(X-1) + W_-(X+1)P_t(X+1) - (W_+(X) + W_-(X))P_t(X), \tag{3}$$

where the transition rates W_+ and W_- for the Schlögl model are given by

$$W_+(X) = k_{+1}a\Omega + k_{-2}bX(X-1)/\Omega, \tag{4}$$
$$W_-(X) = k_{-1}X + k_{+2}X(X-1)(X-2)/\Omega^2, \tag{5}$$

respectively. The stationary solution of the master equation takes the asymptotic form

$$P_{\rm ss}(X) = \exp(-\Omega V_{\rm sto}(\xi) + o(\Omega)) \tag{6}$$

for large Ω. We refer to $V_{\rm sto}(\xi)$ as a stochastic potential. We calculate $V_{\rm sto}(\xi)$ according to

$$\frac{dV_{\rm sto}(\xi)}{d\xi} = \ln\left(\frac{w_-(\xi)}{w_+(\xi)}\right) \tag{7}$$

with normalized transition rates

$$w_+(\xi) = k_{+1}a + k_{-2}b\xi^2, \tag{8}$$
$$w_-(\xi) = k_{-1}\xi + k_{+2}\xi^3. \tag{9}$$

As stochastic effects on the time evolution of the concentration ξ become weaker for larger Ω [24], a system with large Ω is identified as a weakly stochastic system, which is the target of this Letter.

The stationary concentration $\xi_{\rm ss}$ of the weakly stochastic system is defined as the expectation value of ξ by first taking the limit $t \to \infty$ in (3), and then considering the limit $\Omega \to \infty$ in (6). Note that $\xi_{\rm ss}$ minimizes the stochastic potential $V_{\rm sto}(\xi)$ given by (7). In contrast, one can also consider the case of first taking the limit $\Omega \to \infty$ in (3). The concentration dynamics then follows the deterministic rate equation described by

$$\frac{d\xi}{dt} = -k_{+2}\xi^3 + k_{-2}b\xi^2 - k_{-1}\xi + k_{+1}a. \tag{10}$$

The right-hand side of the rate equation is written as a potential force $-dV_{\rm det}/d\xi$, where

$$V_{\rm det}(\xi) = \frac{1}{4}k_{+2}\xi^4 - \frac{1}{3}k_{-2}b\xi^3 + \frac{1}{2}k_{-1}\xi^2 - k_{+1}a\xi. \tag{11}$$

We refer to $V_{\rm det}(\xi)$ as a deterministic potential. Using (11), we find that the rate equation with some parameter values has two stable stationary solutions ξ_- and ξ_+,

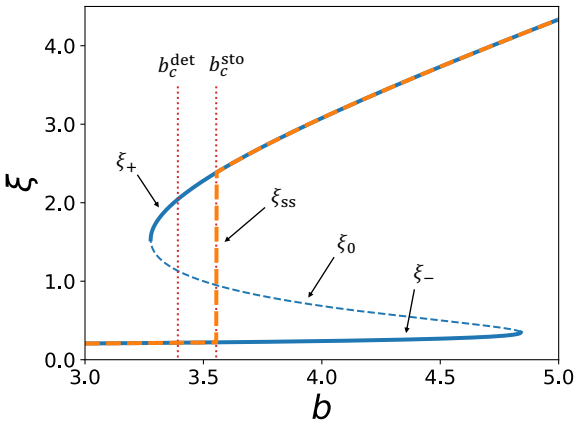


FIG. 2. Relationship between ξ_+, ξ_- and $\xi_{\rm ss}$. In the weakly stochastic system, $\xi_{\rm ss}$ is equal to either ξ_+ or ξ_-, and it is determined as the minimum point of the stochastic potential $V_{\rm sto}(\xi)$ for each b.

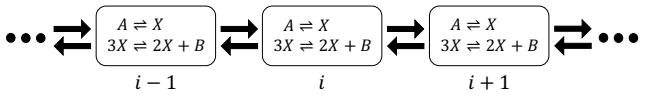


FIG. 3. Schematic figure of a stochastic reaction-diffusion system.

$\xi_- < \xi_+$, at which $V_{\rm det}(\xi)$ becomes local minima. Note that the stationary concentration $\xi_{\rm ss}$ is equal to either ξ_- or ξ_+ as shown in Fig. 2 [22].

Each of the two potentials $V_{\rm sto}$ and $V_{\rm det}$ has two local minima for some parameter values. Below, we focus on this case with changing b while the other parameters are fixed. We define $b_c^{\rm sto}$ as the value at which the two local minimum values of $V_{\rm sto}$ coincide. Similarly, we can define $b_c^{\rm det}$ from $V_{\rm det}$. Note that, in general, $b_c^{\rm sto} \neq b_c^{\rm det}$. For later convenience, let $\kappa_\pm$ be the rate of relaxation to $\xi_\pm$, which provides the characteristic time of the chemical reaction. Explicitly, we have $\kappa_\pm \equiv |V''_{\rm det}(\xi_\pm)| = k_{+2}(\xi_\pm - \xi_0)(\xi_\pm - \xi_\mp)$, where ξ_0 is the local maximum point of $V_{\rm det}(\xi)$.

We now introduce a stochastic reaction-diffusion system [25]. As shown in Fig. 3, $2N+1$ reaction vessels are arranged in the x direction. Let Δx be the length of each vessel in the x direction and $\mathcal{S}$ be the area of the cross-section perpendicular to the x direction. The volume of each vessel is given by $\Omega \equiv \mathcal{S}\Delta x$. The numbers of particles A and B are fixed as the same values for all vessels unless otherwise mentioned later. Each vessel is assumed to be well mixed, and let X_i be the particle number of the i-th vessel, where $-N \le i \le N$. The state of the system is described by $\boldsymbol{X} \equiv (X_{-N}, \cdots, X_N)$, a collection of the particle numbers. Here, we assume that chemical reactions occur only between particles in the same vessel and that particles hop to adjacent vessels. All chemical reactions and particle hoppings are described by the following Markov jump processes. First,

for any i, state transitions associated with chemical reactions in the i-th vessel, $X_i \to X_i \pm 1$, occur at the transition rate $W_\pm(X_i)$. Second, state transitions associated with particle hoppings starting from the i-th vessel, $(X_i, X_{i+\sigma}) \to (X_i - 1, X_{i+\sigma} + 1)$ with $\sigma \in \{1, -1\}$, occur at the transition rate rX_i. The constant r represents the hopping rate per one particle. These transitions correspond to diffusion processes. Note that particles in the vessels at either end hop inward only.

The aim of this Letter is to derive the phase coexistence condition for the weakly stochastic reaction-diffusion system in the limit $N \to \infty$. Here, a phase coexistence state is defined as the spatially inhomogeneous steady state connecting two locally stable stationary concentrations. In particular, we consider two limiting cases, namely (i) the high-hopping-rate regime in which $r/\kappa_\pm \gg 1$ and (ii) the low-hopping-rate regime in which $r/\kappa_\pm \ll 1$. For both regimes, we first take $N \to \infty$ and then $t \to \infty$ and we finally take $\Omega \to \infty$.

Before studying the two limiting cases, we consider the limit $\Omega \to \infty$ in the weakly stochastic reaction-diffusion system. We define $\phi_i(t) \equiv \langle X_i \rangle_t / \Omega$, where $\langle \cdots \rangle_t$ represents the expectation for the stochastic process. Then, in the limit $\Omega \to \infty$, the time evolution of $\phi_i(t)$ is described by [25, 26]

$$\partial_t \phi_i = -k_{+2}\phi_i^3 + k_{-2} b \phi_i^2 - k_{-1}\phi_i + k_{+1} a + r(\phi_{i+1} - 2\phi_i + \phi_{i-1}). \quad (12)$$

Here, we consider the stationary solution of the deterministic equation by taking the limit $t \to \infty$. If the stationary solution is uniquely determined, the order of the two limits $\Omega \to \infty$ and $t \to \infty$ can be exchanged. Therefore, to determine the phase coexistence condition in the limit $N \to \infty$, we first investigate the uniqueness of the stationary solution.

High hopping rate regime.— For the diffusion constant defined by $D \equiv r(\Delta x)^2$, the length reached by the diffusion during the characteristic chemical reaction time $\kappa_\pm^{-1}$ is given by $\ell_\pm^d \equiv \sqrt{D/\kappa_\pm}$ [30]. The length is referred to as the Kuramoto length [31]. Then, the condition $r \gg \kappa_\pm$ implies $\ell_\pm^d \gg \Delta x$. In this case, the continuum description using $\phi(x,t) \equiv \phi_i(t)$ with $x = i\Delta x$ becomes the reaction-diffusion equation

$$\partial_t \phi = -k_{+2}\phi^3 + k_{-2} b \phi^2 - k_{-1}\phi + k_{+1} a + D\partial_x^2 \phi, \quad (13)$$

which provides an asymptotically correct prediction for the solution of (12). Using $L \equiv N\Delta x$, $\phi(x,t)$ is defined in a one-dimensional space $x \in [-L, L]$. As $N \to \infty$ is equal to $L/\ell_\pm^d \to \infty$, phase coexistence states are expressed by the stationary solution satisfying the boundary conditions $\phi(x = -L) = \xi_-$ and $\phi(x = L) = \xi_+$ in the limit $L/\ell_\pm^d \to \infty$. We find the unique stationary solution in this case, and we thus conjecture reasonably that (12) possesses the unique stationary solution under the boundary conditions $\lim_{N\to\infty} \phi_{-N} = \xi_-$ and $\lim_{N\to\infty} \phi_N = \xi_+$ when $r/\kappa_\pm \gg 1$. We thus exchange the order of the limits $\Omega \to \infty$ and $t \to \infty$ and conclude that the phase coexistence condition is given by the point where the deterministic potential $V_{\rm det}(\xi)$ takes the same value at the two minima [26]. Therefore, $b = b_c^{\rm det}$ is the phase coexistence condition for case (i).

We confirm the conjecture through direct numerical simulations of the stochastic reaction-diffusion system using the Gillespie algorithm [32]. As an example, chemical reaction constants are assumed as $k_{-1} = 3.0$, and $k_{+2} = k_{-2} = 1.0$, where $\kappa_+ = \kappa_- = 1.67$ at $b = b_c^{\rm det}$. To satisfy the condition $r \gg \kappa_\pm$, we choose $r = 5.0$. Other parameter values are set to $N = 75$ and $\Delta x = 1.0$. As an initial condition, $X_i(0)/\Omega = \xi_-$ for $i < 0$, $X_i(0)/\Omega = \xi_+$ for $i > 0$, and $X_0(0)/\Omega = \xi_0$. To numerically determine the phase coexistence condition, we measure the interface position $\hat{z}(t)$ at time t, where $\hat{z}(t)$ is given by the x-coordinate at which the concentration profile $\boldsymbol{X}(t)$ crosses the reference concentration $(\xi_+ + \xi_-)/2$. We then estimate the interface velocity $v(b)$ as the statistical average of the displacement of the interface between $t = t_0$ and $t = t_1$:

$$v(b) = \frac{\langle \hat{z}(t_1) - \hat{z}(t_0) \rangle}{t_1 - t_0}, \quad (14)$$

where t_0 is much larger than the relaxation time $\kappa_\pm^{-1}$ and $t_1 - t_0$ is large enough to detect the average velocity. Specifically, we set $t_0 = 10$ and $t_1 = 100$, and the expectation value is estimated by making 100 independent calculations of $\hat{z}(t)$. The phase coexistence condition $b = b_c(\Omega)$ for the system with finite Ω is determined by $v(b_c(\Omega)) = 0$. In Fig. 1, we show the steady state profile ϕ_i for $r = 5.0$ and $\Omega = 100$. Figure 4 shows the Ω dependence of $b_c(\Omega)$. It is observed that $b_c(\Omega)$ approaches $b_c^{\rm det}$ as Ω increases. Such a difference between $b_c(\Omega)$ and $b_c^{\rm det}$ was reported in Ref. [19]. More quantitatively, we numerically find that $b_c(\Omega) - b_c^{\rm det} \simeq 1/\Omega$. Similar behaviors are observed for $r = 0.5$ and $r = 1.0$ [26]. This leads to a conjecture that $b_c(\Omega) \to b_c^{\rm det}$ as $\Omega \to \infty$ for r in a certain range $[r_c, \infty]$. In addition, the numerical result suggests that r_c is smaller than $\kappa_\pm$, which is not understood from a theoretical viewpoint.

Low hopping rate regime.— Next, we consider the low-hopping-rate regime satisfying $r \ll \kappa_\pm$. It has been shown that, for a model similar to ours, an equation corresponding to (12) has many stationary solutions in the low-hopping-rate regime [33]. Therefore, to determine the phase coexistence condition, we first have to consider the limit $t \to \infty$; i.e., we have to deal with the original stochastic process. We thus focus on the stationary solution of the master equation with large Ω. This solution takes the asymptotic form

$$P_{\rm ss}(\boldsymbol{X}) = \exp(-\Omega \tilde{V}_{\rm sto}(\boldsymbol{\xi}; b) + o(\Omega)), \quad (15)$$

where $\boldsymbol{\xi} = (\xi_{-N}, \cdots, \xi_N)$ with $\xi_i \equiv X_i/\Omega$. The most probable configuration minimizes the potential $\tilde{V}_{\rm sto}(\boldsymbol{\xi}; b)$. When this configuration is not homogeneous in space for a certain value of the parameter b, we identify the phase coexistence at the point $b = b_c^{\rm low}$.

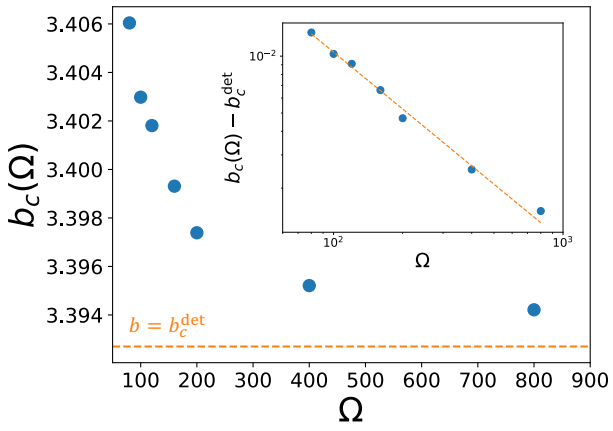


FIG. 4. Ω dependence of $b_c(\Omega)$ for $r = 5.0$. The inset shows a log-log plot with the guide line $b_c(\Omega) - b_c^{\rm det} = \alpha/\Omega$, where $\alpha = 1.05$.

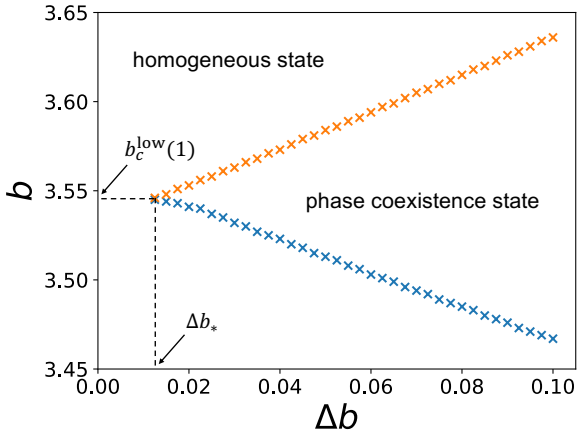


FIG. 5. Phase diagram in the $(b, \Delta b)$ space for $N = 1$.

To extract the phase coexistence condition in the limit $N \to \infty$, we consider a ramped system in which

$$b_i = b + \frac{i}{N}\Delta b \equiv b + \delta b_i \tag{16}$$

is defined for the i-th vessel. The phase coexistence is more likely to occur in the ramped system than in the original system. The phase coexistence condition in the original system can be obtained by considering the limit $N \to \infty$ and $\Delta b \to 0$.

Noting that the hopping rate r is small, we formulate a perturbation theory for calculating the potential $\tilde{V}_{\rm sto}(\boldsymbol{\xi}; b, \Delta b)$ in the ramped system. Ignoring $o(r)$ terms and $o(\Delta b)$ terms in the limit $r \to 0$ and $\Delta b \to 0$, we write the leading order form of the potential as

$$\tilde{V}_{\rm sto}(\boldsymbol{\xi}; b, \Delta b) = \sum_{i=-N}^{N} V_{\rm sto}(\xi_i; b) + \sum_{i=-N}^{N} \partial_b V_{\rm sto}(\xi_i; b) \cdot \delta b_i + rJ(\boldsymbol{\xi}), \tag{17}$$

where $V_{\rm sto}(\xi_i; b)$ is the stochastic potential given by (7) explicitly indicating the b dependence. Substituting (17) into the master equation of the ramped system, we obtain the equation for $J(\boldsymbol{\xi})$ as [26]

$$\sum_{i=-N}^{N} (w_+(\xi_i) - w_-(\xi_i))\partial_{\xi_i} J = \sum_{i=-N}^{N-1} [\xi_i(g_i(\boldsymbol{\xi}) - 1) + \xi_{i+1}(g_i(\boldsymbol{\xi})^{-1} - 1)] \tag{18}$$

with $g_i(\boldsymbol{\xi}) = w_+(\xi_i)w_-(\xi_{i+1})/[w_-(\xi_i)w_+(\xi_{i+1})]$. By numerically solving (18), we obtain $J(\boldsymbol{\xi})$. As a result, $\tilde{V}_{\rm sto}(\boldsymbol{\xi}; b, \Delta b)$ is obtained for each configuration $\boldsymbol{\xi}$ [26].

To find the configuration that minimizes $\tilde{V}_{\rm sto}(\boldsymbol{\xi}; b, \Delta b)$, we consider $2N + 2$ configurations $\boldsymbol{\xi}^{(0)}, \boldsymbol{\xi}^{(1)}, \cdots, \boldsymbol{\xi}^{(2N+1)}$ as candidates of the minimizer, where for any integer k satisfying $0 \le k \le 2N+1$, $\xi_i^{(k)} = \xi_-$ for $-N \le i \le N-k$ and $\xi_i^{(k)} = \xi_+$ for $N-k+1 \le i \le N$. Let k_* be an integer such that $\boldsymbol{\xi}^{(k_*)}$ provides the minimum value of $\tilde{V}_{\rm sto}(\boldsymbol{\xi}^{(k)}; b, \Delta b)$ for a given $(b, \Delta b)$. If $k_* \neq 0,\ 2N+1$, an inhomogeneous configuration $\boldsymbol{\xi}^{(k_*)}$ is most probable for the ramped system with b and Δb. In this case, we identify the phase coexistence state. Figure 5 is a phase diagram in the parameter space $(b, \Delta b)$ for $N = 1$. Similar diagrams can be obtained for any finite N. The diagrams show that a positive Δb_* is necessary to observe the phase coexistence state for systems with finite N. The phase coexistence condition for the special value Δb_* is denoted by $b_c^{\rm low}(N)$. We then find that Δb_* approaches 0 following the law $\Delta b_* \simeq 1/N$ [26]. Therefore, in the limit $N \to \infty$, the phase coexistence state can be realized at $b = b_c^{\rm low}(\infty)$ without ramping. This provides the phase coexistence condition for the original stochastic reaction-diffusion system in the low-hopping-rate regime. In Fig. 6, we show the steady state profile ξ_i determined by the above procedure. In contrast to the high hopping rate regime shown in Fig. 1, the phase coexistence profile has a singular interface.

In Fig. 7, $b_c^{\rm low}(N)$ is presented as a function of r for several values of N. It is observed that $b_c^{\rm low}(N)$ for each r converges to a certain value in the limit $N \to \infty$. Interestingly, the convergence value $b_c^{\rm low}(\infty)$ is clearly different from the phase coexistence condition $b_c^{\rm det}$ derived from the reaction-diffusion equation. Moreover, we find that $b_c^{\rm low}(\infty)$ takes the value around $b_c^{\rm sto}$. This means that the results in the low-hopping-rate regime are qualitatively different from that in the high-hopping-rate regime and suggests that a phase transition occurs at some point r_c beyond which the phase coexistence condition is described by the reaction-diffusion equation.

Concluding Remarks.— In this Letter, we calculated the phase coexistence condition for the weakly stochastic reaction-diffusion system. We found that the phase coexistence condition depends on the hopping rate. In the

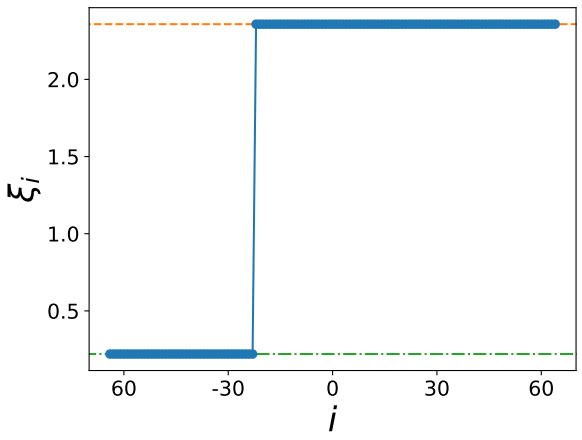


FIG. 6. Phase coexistence state of the stochastic reaction-diffusion system in the low-hopping-rate regime for $r = 0.01$ and $N = 64$. The orange dotted line and green dash-dot line indicate ξ_+ and ξ_-, respectively.

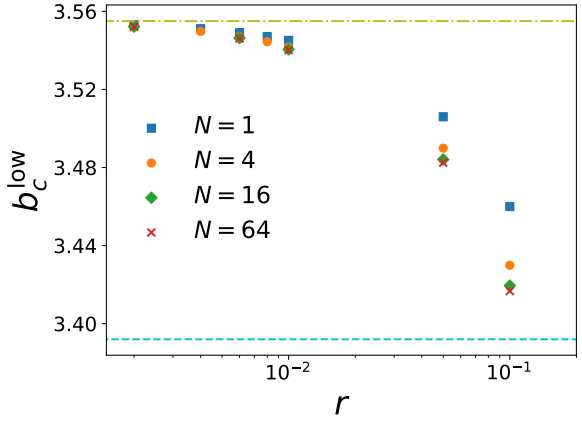


FIG. 7. Hopping rate dependence of the phase coexistence condition $b_c^{\mathrm{low}}(N)$ in the low-hopping-rate regime. In the limit $N \to \infty$, $b_c^{\mathrm{low}}(N)$ converges to a certain value that is clearly different from b_c^{det} shown by the blue dashed line. The yellow dash-dot line shows the value of b_c^{sto}.

high-hopping-rate regime, the phase coexistence is described by the deterministic reaction-diffusion equation and thus determined by the deterministic potential. In contrast, the phase coexistence in the low-hopping-rate regime is determined by a perturbation from the stochastic potential.

To the best of our knowledge, the latter type of phase coexistence has never been observed in reaction-diffusion systems as these systems have typically been studied using macroscopic setups [34]. One possible way to observe such phase coexistence is to use an assemble of bacteria. One can consider a bistable chemical reaction in a cell of each bacterium. If the chemical species are controlled to flow out of the cell, the diffusion-coupled chemical reaction can be designed by preparing many bacteria. Indeed, such a system has been studied for stochastic pattern formation [35]. It would be interesting to observe phase coexistence in the low-hopping-rate regime [26], while a new theoretical method should be devised for analyzing such complex chemical reactions.

From the theoretical viewpoint, the most important problem is to determine whether a phase transition occurs as a function of r. To solve the problem, we need to study the phase coexistence condition for any hopping rate r. As one approach, one can study the Hamilton-Jacobi equation for determining the most probable configuration, which is a natural generalization of our formulation (18) [36–38]. If a singular behavior of $b_c(r)$ is concluded, the two limiting cases reported in this Letter could characterize the different phases. This is left as future work.

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