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# Two Langevin equations in the Doi–Peliti formalism

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## Abstract

A system-size expansion method is incorporated into the Doi–Peliti formalism for stochastic chemical kinetics. The basic idea of the incorporation is to introduce a new decomposition of unity associated with the so-called Cole–Hopf transformation. This approach elucidates a relationship between two different Langevin equations; one is associated with a coherent-state path-integral expression and the other describes density fluctuations. A simple reaction scheme  $X \rightleftharpoons X + X$  is investigated as an illustrative example.

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## 1. Introduction

System-size expansion methods were developed in the analysis of master equations for stochastic chemical kinetics [1–4]. In these methods, under the assumption of an extensive property of macroscopic quantities, fluctuation effects are systematically taken into account by assuming the inverse of a dimensionless system size to be a small perturbation parameter. The leading-order description of a system is provided as a deterministic equation for a density variable due to the law of large numbers, and macroscopic fluctuations are described by a Langevin equation which reduces to the deterministic equation in the limit of an infinite system size.

The Doi–Peliti formalism is known as another standard technique, where master equations are equivalently expressed in terms of a set of creation and annihilation operators [5, 6]. Since the original proposal, many models have been analyzed within this formalism, as seen in a list

of papers quoted in [7]. In particular, a coherent-state path-integral expression is useful for theoretical study, because it takes a form similar to that for a continuum field theory [7–9].

Then, in some cases including an example discussed in this paper, the dynamical action of a coherent-state path-integral expression becomes equivalent to that of a Langevin equation. However, since this Langevin equation does not coincide with that obtained by system-size expansion methods, it does not directly describe the stochastic time evolution of a density. Note that this difference is not well recognized; indeed, as presented in a review paper [10], a Langevin equation obtained by a coherent-state path-integral expression [11] has been analyzed in the problems of fluctuating front propagation of some reaction–diffusion systems as if it describes the time evolution of a density. (See however a different argument in [12].)

In order to resolve this confusing situation, in the present paper, we develop a system-size expansion method for a coherent-state path-integral expression in the Doi–Peliti formalism. Our consideration consists of three steps. In the first place, we express statistical quantities in terms of a variable that is straightforwardly introduced in the coherent-state path-integral expression. In addition to the presentation of such an expression, which was already discussed in [9, 11], we shall provide a clear argument on the indirect correspondence between density fluctuations and trajectories of the Langevin equation associated with the coherent-state path-integral expression. In the second step, we seek a simple expression of density fluctuations in terms of coherent states. Although it has been argued that such an expression would be realized by a so-called Cole–Hopf transformation [13–16], the description involves non-transparent procedures, as will be indicated in the beginning of section 4. Our main idea in the present paper is to introduce a new decomposition of unity associated with the Cole–Hopf transformation, by which we obtain a new path-integral expression. Finally, this expression enables us to perform a system-size expansion, and it directly leads to the Langevin equation describing density fluctuations.

This paper is organized as follows. In section 2, we introduce a model that we study within the Doi–Peliti formalism. In order to make our presentation as instructive and transparent as possible, we focus on a simple reaction scheme  $X \rightleftharpoons X + X$  in a single box under an assumption that diffusion processes are sufficiently fast. In section 3, we review a coherent-state path-integral expression, and derive the first type Langevin equation. In section 4, we propose a new decomposition of unity, by which we derive the second type Langevin equation. In section 5, we briefly discuss the effects of diffusion.

## 2. Model

Let  $\Omega$  be a volume of a box in which particles are confined. We consider a chemical reaction  $X + X \rightleftharpoons X$ ; the duplication reaction  $X \rightarrow X + X$  occurs with the ratio  $\alpha$  for each particle, and its backward reaction  $X + X \rightarrow X$  occurs with the ratio  $\beta$  when two particles are close to each other with less than a reaction length  $r_0$ . We assume that a diffusion time is much smaller than typical time scales of the reaction:  $\alpha^{-1}$  and  $\beta^{-1}$ , and ignore the effect of diffusion for the time being (we will briefly discuss it in section 5). Then, since the probability of finding another particle within a reaction region for a given particle is  $4\pi r_0^3/(3\Omega)$ , the rate of the reaction  $X + X \rightarrow X$  for each pair of particles becomes  $\beta 4\pi r_0^3/(3\Omega)$ . In the argument below, a dimensionless quantity  $4\pi r_0^3/(3\Omega)$  is replaced with  $1/\Omega$ , for notational simplicity. Now, let  $P_n(t)$  be a probability of finding  $n$  particles at time  $t$ . The time evolution of  $P_n(t)$  is described by the master equation

$$\frac{\partial}{\partial t} P_n(t) = \alpha[(n-1)P_{n-1}(t) - nP_n(t)] + \frac{\beta}{\Omega}[n(n+1)P_{n+1}(t) - n(n-1)P_n(t)], \quad (1)$$

where  $P_{-1}(t) \equiv 0$ .

The state of the system is specified by a vector  $|n\rangle$ , which represents a situation that there are  $n$  particles in the system. Let  $V$  be a vector space spanned by an orthogonal set:  $V = \{|n\rangle | n = 0, \dots, \infty\}$ . A remarkable idea of the Doi–Peliti formalism [5, 6] is that a series of infinite number of  $P_n(t)$  is collectively treated as a single vector in  $V$ :

$$|\psi(t)\rangle = \sum_{n=0}^{\infty} P_n(t)|n\rangle. \tag{2}$$

Then, the master equation (1) is equivalently rewritten in a compact form

$$\frac{\partial}{\partial t}|\psi(t)\rangle = -\hat{H}|\psi(t)\rangle, \tag{3}$$

where  $\hat{H}$  is an infinite-dimensional matrix. This matrix is expressed in terms of two matrices  $\hat{a}$  and  $\hat{a}^\dagger$  defined by  $\hat{a}|n+1\rangle = (n+1)|n\rangle$  and  $\hat{a}^\dagger|n\rangle = |n+1\rangle$  for any  $n$  (with  $\hat{a}|0\rangle = 0$ ):

$$\hat{H} = -\alpha(\hat{a}^\dagger - 1)\hat{a}^\dagger\hat{a} - \frac{\beta}{\Omega}(1 - \hat{a}^\dagger)\hat{a}^\dagger\hat{a}^2. \tag{4}$$

The two matrices satisfy the bosonic commutation relations  $[\hat{a}, \hat{a}^\dagger] = 1$  and  $[\hat{a}, \hat{a}] = [\hat{a}^\dagger, \hat{a}^\dagger] = 0$ . Since  $\hat{a}^\dagger$  and  $\hat{a}$  are similar to the creation and annihilation operators, and  $\hat{H}$  is an analog of the Hamiltonian, the Doi–Peliti formalism is also called the second-quantization method.

For any  $|v_1\rangle$  and  $|v_2\rangle$  in  $V$ , the inner product  $\langle v_1|v_2\rangle$  is naturally defined by noting  $\langle m|n\rangle = n!\delta_{m,n}$ . There is a special vector  $|\mathcal{P}\rangle$  such that  $\langle n|\mathcal{P}\rangle = 1$  for any  $n$ , which is employed to express the expectation value of observables. This vector is explicitly written as  $|\mathcal{P}\rangle = e^{\hat{a}^\dagger}|0\rangle$ . Let  $A(x)$  be a polynomial of a variable  $x$ . We consider the expectation value of  $A(n)$  at time  $t = \tau$ , which is denoted by  $\langle A(n(\tau))\rangle$ . By using the vector  $|\mathcal{P}\rangle$ , we write  $\langle A(n(\tau))\rangle \equiv \sum_{n=0}^{\infty} A(n)P_n(\tau) = \langle \mathcal{P}|A(\hat{a}^\dagger\hat{a})|\psi(\tau)\rangle$ . In order to simplify this expression further, we define a polynomial  $\mathcal{A}(x)$  associated with  $A(x)$  by the relation  $\langle \mathcal{P}|A(\hat{a}^\dagger\hat{a})\rangle = \langle \mathcal{P}|\mathcal{A}(\hat{a})\rangle$ . That is,  $\mathcal{A}(\hat{a})$  is obtained by the *normal ordering* of  $A(\hat{a}^\dagger\hat{a})$ . As an example, we demonstrate for the simplest non-trivial case  $A(x) = x^2$ . Since  $\langle \mathcal{P}|(\hat{a}^\dagger\hat{a})^2\rangle = \langle \mathcal{P}|\hat{a}\hat{a}^\dagger\hat{a} = \langle \mathcal{P}|(\hat{a}^2 + \hat{a})\rangle$ , one finds  $\mathcal{A}(x) = x^2 + x$ . More generally, by mathematical induction, one can prove that  $\mathcal{A}(x) = x^k$  for  $A(x) = x(x-1)\cdots(x-k+1)$  with any integer  $k$ . The final expression of the expectation value of  $A(n)$  at  $t = \tau$  becomes

$$\langle A(n(\tau))\rangle = \langle \mathcal{P}|\mathcal{A}(\hat{a})|\psi(\tau)\rangle. \tag{5}$$

### 3. Coherent-state path-integral expression

With the ‘second-quantized’ expression of the master equation (3), it is quite natural to consider the path-integral form of the expectation value (5). In this section, we explain a procedure to obtain a path-integral expression by using coherent states within the model (4). The result was already obtained in [11], but we shall provide a slightly different presentation of the derivation.

Coherent states are eigenstates of the matrix  $\hat{a}$ . They are in general written as

$$|z\rangle \equiv e^{z\hat{a}^\dagger}|0\rangle \tag{6}$$

with a complex number parameter  $z$  being the eigenvalue:  $\hat{a}|z\rangle = z|z\rangle$ . The corresponding bra vector is denoted by  $\langle z|$ , that is,  $\langle z| = \langle 0|e^{z^*\hat{a}}$ , where  $z^*$  is the complex conjugate of  $z$ . The following decomposition of unity plays an essential role in constructing a path-integral expression with the coherent states:

$$\mathbf{1} = \sum_{n=0}^{\infty} \frac{1}{n!}|n\rangle\langle n| = \sum_{n=0}^{\infty} \sum_{m=0}^{\infty} \frac{1}{n!}|n\rangle\langle m|\delta_{m,n} = \int \frac{d^2z}{\pi} e^{-|z|^2}|z\rangle\langle z|, \tag{7}$$

where we have used

$$\delta_{n,m} = \int \frac{d^2z}{\pi n!} e^{-|z|^2} z^{*m} z^n, \quad (8)$$

with the integration measure  $d^2z = d(\text{Re } z) d(\text{Im } z)$ . See [7] and [9] for the path-integral expression on the basis of (7).

Here, we note a different decomposition of unity

$$\mathbf{1} = \int_{-\infty}^{\infty} d\phi \int_{-\infty}^{\infty} \frac{d\varphi}{2\pi} |\phi\rangle \langle -i\varphi| e^{-i\phi\varphi}, \quad (9)$$

where we have used the formula

$$\delta_{n,m} = \frac{1}{n!} \int_{-\infty}^{\infty} d\phi \phi^n \left(-\frac{d}{d\phi}\right)^m \delta(\phi) = \frac{1}{n!} \int_{-\infty}^{\infty} d\phi \int_{-\infty}^{\infty} \frac{d\varphi}{2\pi} \phi^n (i\varphi)^m e^{-i\phi\varphi}. \quad (10)$$

Note that  $\phi$  and  $\varphi$  are *real* variables. The path-integral expression on the basis of the decomposition of unity (9) might be less familiar, but we remind readers that this expression was presented in the seminal paper by Peliti [8]. It should be equivalent to the standard one obtained by using (7). Indeed, when we employ the expression with  $(\phi, i\varphi)$ , we can always move to the standard expression by formally replacing  $(\phi, i\varphi)$  with  $(z, z^*)$ . In addition, as far as we understand, the expression obtained by (9) is more transparent in the sense that the integration can be considered in an explicit manner. From these reasons, we review the coherent-state path-integral expression by using (9).

The time integration of (3) yields

$$|\psi(\tau)\rangle = \lim_{\Delta t \rightarrow 0} [\exp(-\hat{H} \Delta t)]^{\tau/\Delta t} |\psi(0)\rangle. \quad (11)$$

We assume that an initial condition obeys a Poisson distribution with its average  $\bar{n}_0$ . The initial state  $|\psi(0)\rangle$  is then expressed as

$$|\bar{n}_0; \text{Pois}\rangle \equiv \sum_{i=0}^{\infty} e^{-\bar{n}_0} \frac{\bar{n}_0^i}{i!} |i\rangle = e^{\bar{n}_0(\hat{a}^\dagger - 1)} |0\rangle. \quad (12)$$

For the moment, we focus on the case

$$\alpha \bar{n}_0 - \frac{\beta}{\Omega} \bar{n}_0^2 > 0, \quad (13)$$

and the other case will be discussed at the end of this section. By inserting the identity (9) into (5) with (11) and (12), we express  $\langle A(n(\tau)) \rangle$  as

$$\begin{aligned} \langle A(n(\tau)) \rangle &= \lim_{\Delta t \rightarrow 0} \left( \prod_{t=0}^{\tau} \int_{-\infty}^{\infty} d\phi_t \int_{-\infty}^{\infty} \frac{d\varphi_t}{2\pi} \right) \langle \mathcal{P} | \mathcal{A}(\hat{a}) | \phi_t \rangle \\ &\times \left[ \prod_{t=\Delta t}^{\tau} \langle -i\varphi_t | e^{-\hat{H} \Delta t} | \phi_{t-\Delta t} \rangle e^{-i\phi_t \varphi_t} \right] e^{-i\varphi_0 \phi_0} \langle -i\varphi_0 | \bar{n}_0; \text{Pois} \rangle, \end{aligned} \quad (14)$$

where the time index  $t$  runs from zero to  $\tau$  in steps of  $\Delta t$ . (We assume  $\tau = N \Delta t$  with the integer  $N$ .) Since each term of  $\hat{H}$  in (4) is arranged in normal order, we derive

$$\langle -i\varphi_t | e^{-\hat{H} \Delta t} | \phi_{t-\Delta t} \rangle = \langle -i\varphi_t | \phi_{t-\Delta t} \rangle \exp(-H(i\varphi_t, \phi_{t-\Delta t}) \Delta t), \quad (15)$$

where  $H(i\varphi_t, \phi_{t-\Delta t})$  is defined by simply replacing  $\hat{a}^\dagger$  and  $\hat{a}$  in  $\hat{H}$  with  $i\varphi_t$  and  $\phi_{t-\Delta t}$ , respectively. We also have  $\langle -i\varphi_t | \phi_{t-\Delta t} \rangle = \exp(i\varphi_t \phi_{t-\Delta t})$ ,  $\langle \mathcal{P} | \mathcal{A}(\hat{a}) | \phi_\tau \rangle = \langle 1 | \phi_\tau \rangle \mathcal{A}(\phi_\tau) =$

$\mathcal{A}(\phi_\tau) e^{\phi_\tau}$  and  $\langle -i\varphi_0 | \bar{n}_0; \text{Pois} \rangle = \exp(i\varphi_0 \bar{n}_0 - \bar{n}_0)$ . By substituting these results into (14), we obtain a path-integral expression

$$\langle A(n(\tau)) \rangle = \lim_{\Delta t \rightarrow 0} \left( \prod_{t=0}^{\tau} \int_{-\infty}^{\infty} d\phi_t \int_{-\infty}^{\infty} \frac{d\varphi_t}{2\pi} \right) \mathcal{A}(\phi_\tau) \exp[-S(\{i\varphi\}, \{\phi\})], \quad (16)$$

where the ‘action’  $S$  is calculated as

$$S(\{i\varphi\}, \{\phi\}) = -\phi_\tau + i\varphi_0(\phi_0 - \bar{n}_0) + \bar{n}_0 + \sum_{t=\Delta t}^{\tau} \Delta t \left[ i\varphi_t \frac{\phi_t - \phi_{t-\Delta t}}{\Delta t} - \alpha((i\varphi_t)^2 - i\varphi_t)\phi_{t-\Delta t} - \frac{\beta}{\Omega}(i\varphi_t - (i\varphi_t)^2)\phi_{t-\Delta t}^2 \right]. \quad (17)$$

Although  $S$  is a complex valued functional as it is, the integration with respect to  $\varphi_t$  in (16) yields a real valued functional of  $\phi$ . Here, the integration of  $\varphi_t$  in (16) is carried out from  $-\infty$  to  $+\infty$  along the real axis. We assume that the integration path can be shifted to the straight line from  $-\infty - i$  to  $+\infty - i$ , keeping the integration value unchanged. This shift is equivalent to the replacement of  $\varphi_t$  with  $\bar{\varphi}_t - i$ , where  $\bar{\varphi}_t$  is a real variable. Then, the action (17) becomes

$$\tilde{S}(\{i\bar{\varphi}\}, \{\phi\}) = i\bar{\varphi}_0(\phi_0 - \bar{n}_0) + \sum_{t=\Delta t}^{\tau} \Delta t \left[ i\bar{\varphi}_t \frac{\phi_t - \phi_{t-\Delta t}}{\Delta t} - \alpha((i\bar{\varphi}_t)^2 + i\bar{\varphi}_t)\phi_{t-\Delta t} + \frac{\beta}{\Omega}(i\bar{\varphi}_t + (i\bar{\varphi}_t)^2)\phi_{t-\Delta t}^2 \right]. \quad (18)$$

Next, we linearize the terms quadratic in  $\bar{\varphi}_t$  by introducing an auxiliary real variable  $y_t$ :

$$\exp \left[ (i\bar{\varphi}_t)^2 \left( \alpha\phi_{t-\Delta t} - (\beta/\Omega)\phi_{t-\Delta t}^2 \right) (\Delta t) \right] = \int_{-\infty}^{\infty} \frac{dy_t}{\sqrt{2\pi}} \exp \left[ -\frac{1}{2}y_t^2 + i\bar{\varphi}_t \sqrt{2(\alpha\phi_{t-\Delta t} - (\beta/\Omega)\phi_{t-\Delta t}^2)} (\Delta t)^{1/2} y_t \right]. \quad (19)$$

Note that this identity is valid only when

$$\alpha\phi_{t-\Delta t} - (\beta/\Omega)\phi_{t-\Delta t}^2 \geq 0. \quad (20)$$

We conjecture that trajectories that do not satisfy this condition do not contribute the path-integration in (16) under condition (13). See the discussion below (24). Finally, by employing the Fourier transformation formula of Dirac’s delta function, we can rewrite (16) as

$$\langle A(n(\tau)) \rangle = \lim_{\Delta t \rightarrow 0} \left( \prod_{t=\Delta t}^{\tau} \int_{-\infty}^{\infty} d\phi_t \int_{-\infty}^{\infty} \frac{dy_t}{\sqrt{2\pi}} e^{-\frac{1}{2}y_t^2} \right) \mathcal{A}(\phi_\tau) \left[ \prod_{t=\Delta t}^{\tau} \delta(\Phi(\phi_t, \phi_{t-\Delta t}, y_t)) \right]_{\phi_0=\bar{n}_0}, \quad (21)$$

where

$$\Phi(\phi_t, \phi_{t-\Delta t}, y_t) = \phi_t - \phi_{t-\Delta t} - \left( \alpha\phi_{t-\Delta t} - \frac{\beta}{\Omega}\phi_{t-\Delta t}^2 \right) (\Delta t) - \sqrt{2 \left( \alpha\phi_{t-\Delta t} - \frac{\beta}{\Omega}\phi_{t-\Delta t}^2 \right)} (\Delta t)^{1/2} y_t. \quad (22)$$

The equation  $\Phi = 0$  in the limit  $\Delta t \rightarrow 0$  is expressed as

$$\frac{d}{dt}\phi = \alpha\phi - \frac{\beta}{\Omega}\phi^2 + \sqrt{2 \left( \alpha\phi - \frac{\beta}{\Omega}\phi^2 \right)} \cdot \eta, \quad (23)$$

where the quantity  $\eta(t)$  defined as the continuum limit of  $y_t/\sqrt{\Delta t}$  is a white Gaussian noise that satisfies  $\langle \eta(t)\eta(t') \rangle = \delta(t - t')$ , and the symbol  $\cdot$  in front of  $\eta$  represents the product in the sense of Itô. Therefore, formula (21) with (22) is written as

$$\langle A(n(\tau)) \rangle = \langle \mathcal{A}(\phi(\tau)) \rangle_\phi, \quad (24)$$

where  $\langle \cdot \rangle_\phi$  represents the expectation value over noise sequences of the Langevin equation (23). It should be noted that trajectories  $\phi(t)$  generated by (23) with the initial condition  $\phi(0) = \bar{n}_0$  satisfy  $\alpha\phi - (\beta/\Omega)\phi^2 \geq 0$  for any time  $t$  as discussed in [11]. Then, one may obtain (18) starting from (23). This implies that the conjecture mentioned below (20) is valid.

We choose  $A(n) = n/\Omega$  in (24) as the simplest example. By setting  $u(t) = \phi(t)/\Omega$ , we have

$$\left\langle \frac{n(\tau)}{\Omega} \right\rangle = \langle u(\tau) \rangle_u, \quad (25)$$

where  $\langle \cdot \rangle_u$  represents the expectation value over noise sequences of the Langevin equation

$$\frac{d}{dt}u = \alpha u - \beta u^2 + \sqrt{\frac{2(\alpha u - \beta u^2)}{\Omega}} \cdot \eta. \quad (26)$$

Surprisingly, the average behavior of the density  $n/\Omega$  is exactly described by the Langevin equation (26) for any  $\Omega$ . The Langevin equation (26) supplemented with a diffusion term is called the stochastic Fisher–Kolmogorov–Petrovsky–Piscounov (s-FKPP) equation, which appears in several research fields [10] (for example, see [17] for applications to the problems of high-energy hadron scattering). Historically, the s-FKPP equation for the chemical reaction  $X \rightleftharpoons X + X$  was derived on the basis of the coherent-state path-integral expression with the complex parameterization  $(z, z^*)$  [11] in studying non-trivial noise effects for front-propagation [18, 19]. Here, it is worthwhile noting that  $\phi$  does *not* correspond to the number of particles, as we can see the difference between the two polynomials  $A$  and  $\mathcal{A}$  that appear in (24). It is only when we take the simplest case  $A(x) = x$  discussed in (25) that  $\mathcal{A}(x)$  becomes equivalent to  $A(x)$ . Therefore, one must *not* interpret that (26) is the equation for a fluctuating density. Indeed, (26) is different from that obtained by system-size expansion methods for the master equation (1). (Compare (26) with (50) derived in the next section.)

At the end of this section, we address three technical remarks. First, we discuss the validity of the assumption mentioned above (18). We do not have a mathematical proof for the claim that the integration path can be shifted as such described there. However, this is plausible because we can prove the result (24) in the following manner. It is sufficient to consider the case where  $A(x)$  is given by  $A_k(x) = x(x - 1) \cdots (x - k + 1)$  with an arbitrary positive integer  $k$ . The corresponding polynomial  $\mathcal{A}(x)$  becomes  $\mathcal{A}_k(x) = x^k$ . By taking the  $k$ th derivative of the identity

$$\sum_{n=0}^{\infty} \frac{s^n \bar{n}_0^n}{n!} = e^{\bar{n}_0 s} \quad (27)$$

with respect to  $s$ , and setting  $s = 1$ , we obtain

$$\sum_{n=0}^{\infty} \frac{\bar{n}_0^n e^{-\bar{n}_0}}{n!} n(n - 1) \cdots (n - k + 1) = \bar{n}_0^k, \quad (28)$$

which means

$$\langle A_k(n(0)) \rangle = \mathcal{A}_k(\bar{n}_0). \quad (29)$$

Next, following the Itô formula, we find from the Langevin equation (23) for  $\phi$

$$\begin{aligned} \frac{d \langle \mathcal{A}_k(\phi) \rangle_\phi}{dt} &= \left\langle \left( \alpha \phi - \frac{\beta}{\Omega} \phi^2 \right) \frac{d \mathcal{A}_k(\phi)}{d\phi} \right\rangle_\phi + \left\langle \left( \alpha \phi - \frac{\beta}{\Omega} \phi^2 \right) \frac{d^2 \mathcal{A}_k(\phi)}{d\phi^2} \right\rangle_\phi \\ &= \alpha \langle k \mathcal{A}_k + k(k-1) \mathcal{A}_{k-1} \rangle_\phi - \frac{\beta}{\Omega} \langle k \mathcal{A}_{k+1} + k(k-1) \mathcal{A}_k \rangle_\phi. \end{aligned} \tag{30}$$

On the other hand, by using the master equation (1), we can confirm that the differential equation for  $d \langle A_k(n(t)) \rangle / dt$  has the structure identical with that of the right-hand side of (30). This observation combined with (29) leads to  $\langle A_k(n(t)) \rangle = \langle \mathcal{A}_k(\phi(t)) \rangle_\phi$  for any  $k$  at any  $t$ . Since arbitrary polynomials can be expressed as a linear combination of  $\{A_k\}$ , (24) has been proved.

Second, when  $\bar{n}_0$  does not satisfy condition (13), the integration formula (19) is not available. Therefore, result (24) with the Langevin equation (23) is valid only for the case (13).

Finally, let us consider two time quantities such as  $\langle A(n(t_1))A(n(t_2)) \rangle$ . In order to calculate such a quantity, we need a path-integral expression of the conditional probability  $P(n, \tau | n_0, 0)$ . If we replace the Poisson initial condition with  $\delta(n - n_0)$ , the path-integral expression in this section becomes complicated.

#### 4. Density fluctuations

In order to have a direct correspondence with density fluctuations in the coherent-state path-integral expression, a nonlinear transformation in the action (17)

$$\phi = \nu e^{-\chi}, \quad i\varphi = e^\chi \tag{31}$$

has been employed [9, 13–15], which is called the Cole–Hopf transformation. (In the standard formulation with  $(z, z^*)$ ,  $(\phi, i\varphi)$  in (31) is simply replaced with  $(z, z^*)$ .) Quite formally, substituting (31) into (17), neglecting terms with third and higher powers of  $\chi$ , expecting cancellation of several terms appearing in contributions at  $t = \tau$ , and avoiding considerations on the integration path, one can obtain the dynamical action of a Langevin equation for the variable  $\nu/\Omega$ . This Langevin equation coincides with that obtained by system-size expansion methods.

Although the final result is plausible, it seems difficult to judge the validity of the procedures. In particular, in the standard formulation with  $(z, z^*)$ , there is no complex number  $\chi$  that would satisfy  $z = \nu e^{-\chi}$  and  $z^* = e^\chi$ , as easily checked for an example  $z = 1 + i$ . In the formulation with  $(\phi, i\varphi)$ , the integration path of  $\chi$  is described by  $\chi = \log |\varphi| + i\pi \operatorname{sgn}(\varphi)/2$  with  $-\infty < \varphi < 0$  and  $0 < \varphi < \infty$ . The calculation after that seems complicated. Toward a justification of (31), recently, an operator version of the Cole–Hopf transformation has been presented [16]; but its mathematical foundation is not obvious.

Based on these understandings, we propose a framework in which the calculation procedures mentioned above are formulated without any mathematical difficulties. Our basic idea is to introduce a new decomposition of unity associated with the Cole–Hopf transformation. As a preparation, motivated by (31), we define

$$\tilde{z} \equiv z/|z| \tag{32}$$

for any non-zero complex number  $z$ . In the argument below, the complex variable  $z$  is always connected to two real variables  $\mu$  and  $\theta$  as

$$z = \mu e^{-i\theta}, \tag{33}$$



where  $\mu \geq 0$  and  $-\pi \leq \theta \leq \pi$ . Then,  $\tilde{z} = e^{-i\theta}$ . By using coherent states parametrized by  $z$  and  $\tilde{z}$ , we find a new decomposition of unity as follows:

$$\begin{aligned} \mathbf{1} &= \sum_{\ell} \frac{1}{\ell!} |\ell\rangle \langle \ell| \\ &= \sum_{\ell} \frac{1}{\ell! \ell!} \int_0^{\infty} d\mu e^{-\mu} \mu^{\ell} |\ell\rangle \langle \ell| \\ &= \sum_{\ell, m} \frac{1}{\ell! m!} \int_0^{\infty} d\mu \int_{-\pi}^{\pi} \frac{d\theta}{2\pi} e^{-\mu} \mu^{\ell} e^{-i(\ell-m)\theta} |\ell\rangle \langle m| \\ &= \int_0^{\infty} d\mu \int_{-\pi}^{\pi} \frac{d\theta}{2\pi} |z\rangle \langle \tilde{z}| e^{-\mu}. \end{aligned} \quad (34)$$

We shall employ this decomposition of unity for constructing a path-integral expression.

We consider the transition probability  $P(n_f, \tau | n_i, 0)$ , which is the probability of finding  $n_f$  particles at time  $t = \tau$  provided that there are  $n_i$  particles at  $t = 0$ . This is expressed as

$$P(n_f, \tau | n_i, 0) = \frac{1}{n_f!} \langle n_f | e^{-\hat{H}\tau} | n_i \rangle. \quad (35)$$

Its path-integral expression on the basis of (34) is written as

$$\begin{aligned} P(n_f, \tau | n_i, 0) &= \lim_{\Delta t \rightarrow 0} \left( \prod_{t=0}^{\tau} \int_0^{\infty} d\mu_t \int_{-\pi}^{\pi} \frac{d\theta_t}{2\pi} \right) \frac{1}{n_f!} \langle n_f | z_{\tau} \rangle \\ &\times \left[ \prod_{t=\Delta t}^{\tau} \langle \tilde{z}_t | e^{-\hat{H}\Delta t} | z_{t-\Delta t} \rangle e^{-\mu_t} \right] e^{-\mu_0} \langle \tilde{z}_0 | n_i \rangle. \end{aligned} \quad (36)$$

(See the appendix for the corresponding path-integral expression in terms of particle numbers instead of coherent states.) As did in the previous section, we have  $\langle \tilde{z}_t | e^{-\hat{H}\Delta t} | z_{t-\Delta t} \rangle = \langle \tilde{z}_t | z_{t-\Delta t} \rangle \exp(-H(\tilde{z}_t^*, z_{t-\Delta t})\Delta t)$ , where  $H$  is expressed as

$$H(\tilde{z}_t^*, z_{t-\Delta t}) = -\alpha((e^{i\theta_t})^2 - e^{i\theta_t})\mu_{t-\Delta t} e^{-i\theta_{t-\Delta t}} - \frac{\beta}{\Omega}(e^{i\theta_t} - (e^{i\theta_t})^2)(\mu_{t-\Delta t})^2 e^{-2i\theta_{t-\Delta t}}. \quad (37)$$

We then note

$$\begin{aligned} \prod_{t=\Delta t}^{\tau} \langle \tilde{z}_t | z_{t-\Delta t} \rangle e^{-\mu_t} &= \prod_{t=\Delta t}^{\tau} \exp(-\mu_t + \mu_{t-\Delta t} e^{i(\theta_t - \theta_{t-\Delta t})}) \\ &= \prod_{t=\Delta t}^{\tau} e^{-\mu_t + \mu_{t-\Delta t} + i\mu_{t-\Delta t}(\theta_t - \theta_{t-\Delta t}) + O(\Delta t^2)} \\ &= e^{-\mu_{\tau} + \mu_0 + i\theta_{\tau}\mu_{\tau} - i\theta_0\mu_0} \prod_{t=\Delta t}^{\tau} e^{-i\theta_t(\mu_t - \mu_{t-\Delta t}) + O(\Delta t^2)}, \end{aligned} \quad (38)$$

where we have used the estimation  $\theta_t - \theta_{t-\Delta t} \simeq O(\Delta t)$ . Furthermore, we have  $\langle n_f | z_{\tau} \rangle = z_{\tau}^{n_f} = \mu_{\tau}^{n_f} e^{-in_f\theta_{\tau}}$  and  $\langle \tilde{z}_0 | n_i \rangle = e^{i\theta_0 n_i}$ . Substitution of these results into (36) yields

$$\begin{aligned} P(n_f, \tau | n_i, 0) &= \lim_{\Delta t \rightarrow 0} \left( \prod_{t=0}^{\tau} \int_0^{\infty} d\mu_t \int_{-\pi}^{\pi} \frac{d\theta_t}{2\pi} \right) e^{-i\theta_0(\mu_0 - n_i) + i\theta_{\tau}(\mu_{\tau} - n_f)} \\ &\times \frac{1}{n_f!} \mu_{\tau}^{n_f} e^{-\mu_{\tau}} \exp[-\mathcal{S}(\{i\theta_t\}, \{\mu_t\})], \end{aligned} \quad (39)$$

with

$$\mathcal{S}(\{i\theta_t\}, \{\mu_t\}) = \sum_{t=\Delta t}^{\tau} [H(\bar{z}_t^*, z_{t-\Delta t})\Delta t + i\theta_t(\mu_t - \mu_{t-\Delta t})]. \quad (40)$$

Here, we perform the integrations with respect to  $\theta_0$  and  $\theta_\tau$ . Noting that the limit  $\Delta t \rightarrow 0$  is taken in the final expression, we obtain

$$P(n_f, \tau | n_i, 0) = \lim_{\Delta t \rightarrow 0} \left( \prod_{t=\Delta t}^{\tau-\Delta t} \int_0^\infty d\mu_t \int_{-\pi}^\pi \frac{d\theta_t}{2\pi} \right) \frac{1}{n_f!} n_f^{n_f} e^{-n_f} e^{-\mathcal{S}(\{i\theta_t\}, \{\mu_t\})} \Big|_{\mu_0=n_i, \mu_\tau=n_f}. \quad (41)$$

This expression is exact and if one simply replaces  $i\theta_t$  and  $\mu_t$  with  $\chi_t$  and  $v_t$ , respectively, the resulting action  $\mathcal{S}(\{\chi_t\}, \{v_t\})$  is equal to the one obtained by a formal procedure with the Cole–Hopf transformation (31). Therefore, we claim that our argument provides a mathematical foundation for the Cole–Hopf transformation.

Now, recalling that  $\Omega$  is a dimensionless volume of the system, we focus on the regime  $n_f \gg 1$  and  $n_i \gg 1$  under the assumption  $\Omega \gg 1$ . More explicitly, by setting  $\rho_i = n_i/\Omega$  and  $\rho_f = n_f/\Omega$ , we assume a large deviation property

$$P(\rho_f \Omega, \tau | \rho_i \Omega, 0) \simeq e^{-\Omega \mathcal{F}(\rho_f, \tau | \rho_i, 0)}, \quad (42)$$

where  $\rho_i$  and  $\rho_f$  are the particle densities at initial and final times and are finite in general. Note that the relation  $A(\Omega) \simeq B(\Omega)$  in this paper means  $(\log A(\Omega) - \log B(\Omega))/\Omega \rightarrow 0$  in the limit  $\Omega \rightarrow \infty$ . The quantity  $\mathcal{F}(\rho_f, \tau | \rho_i, 0)$ , which is called a large deviation function, characterizes fluctuation properties of a density in a macroscopic system. The problem we consider here is to derive a simpler stochastic system that reproduces  $\mathcal{F}(\rho_f, \tau | \rho_i, 0)$  defined in (42).

Since  $\mu_0$  and  $\mu_\tau$  are fixed to  $\rho_i \Omega$  and  $\rho_f \Omega$ , respectively, the dominant contribution of the  $\mu_t$  integration in (41) comes from a region  $\mu_t \sim O(\Omega)$ . On the other hand, since there is the term  $\exp(i\theta_t(\mu_t - \mu_{t-\Delta t}))$  in the integrand, the dominant contribution of the  $\theta_t$  integration comes from a region  $\theta_t \sim O(1/\Omega)$ . From these observations, we make the transformation of variables as  $\rho_t = \mu_t/\Omega$  and  $\pi_t = \Omega\theta_t$  in the path-integral expression (41) so that the integration over a region where  $\rho_t \simeq O(1)$  and  $\pi_t \simeq O(1)$  provides the dominant contribution. We also note the relation  $n_f! \simeq n_f^{n_f} e^{-n_f}$  valid for  $n_f \gg 1$ . We then have

$$e^{-\Omega \mathcal{F}(\rho_f, \tau | \rho_i, 0)} \simeq \lim_{\Delta t \rightarrow 0} \left( \prod_{t=\Delta t}^{\tau-\Delta t} \int_0^\infty d\rho_t \int_{-\infty}^\infty \frac{d\pi_t}{2\pi} \right) e^{-\bar{\mathcal{S}}(\{i\pi_t\}, \{\rho_t\})} \Big|_{\rho_0=\rho_i, \rho_\tau=\rho_f}, \quad (43)$$

where  $\bar{\mathcal{S}}(\{i\pi_t\}, \{\rho_t\})$  is expanded in terms of  $1/\Omega$  and  $\Delta t$  as

$$\bar{\mathcal{S}}(\{i\pi_t\}, \{\rho_t\}) = (\Delta t) \sum_{t=\Delta t}^{\tau} \left[ i\pi_t \Psi_1(\rho_t, \rho_{t-\Delta t}) + \frac{1}{2\Omega} (\pi_t)^2 \Psi_2(\rho_{t-\Delta t}) + R(i\pi_t, \rho_{t-\Delta t}) \right] \quad (44)$$

with

$$\Psi_1(\rho_t, \rho_{t-\Delta t}) = \frac{\rho_t - \rho_{t-\Delta t}}{\Delta t} - (\alpha \rho_{t-\Delta t} - \beta \rho_{t-\Delta t}^2), \quad (45)$$

$$\Psi_2(\rho_{t-\Delta t}) = \alpha \rho_{t-\Delta t} + \beta \rho_{t-\Delta t}^2, \quad (46)$$

$$R(i\pi_t, \rho_{t-\Delta t}) = O\left(\frac{1}{\Omega^2}\right) + O(\Delta t). \quad (47)$$

The  $\pi_t$  integration in (43) with  $R(i\pi_t, \rho_{t-\Delta t})$  in  $\bar{\mathcal{S}}$  being ignored yields the formula

$$\mathcal{F}(\rho_f, \tau | \rho_i, 0) = \lim_{\Delta t \rightarrow 0} \min_{\rho_i \rightarrow \rho_f} (\Delta t) \sum_{t=\Delta t}^{\tau} \frac{[\Psi_1(\rho_t, \rho_{t-\Delta t})]^2}{2\Psi_2(\rho_{t-\Delta t})}, \quad (48)$$

where  $\min_{\rho_i \rightarrow \rho_f}$  represents the minimization with respect to  $\{\rho_t\}$  with the boundary conditions  $\rho_0 = \rho_i$  and  $\rho_\tau = \rho_f$  fixed. If one includes the term  $R(i\pi_t, \rho_{t-\Delta t})$  in  $\tilde{\mathcal{S}}$  and treats it as a perturbation, one can show that it does not contribute to the large deviation function  $\mathcal{F}$ . Therefore, as far as we are concerned with the large deviation property, we may neglect  $R(i\pi_t, \rho_{t-\Delta t})$  in  $\tilde{\mathcal{S}}$ .

Then, by applying the same procedures as (19) and (21) in the previous section to the expression (43) and (44), we arrive at the final result

$$P(\rho_f \Omega, \tau | \rho_i \Omega, 0) \simeq \langle \delta(\rho(\tau) - \rho_f) \rangle_\rho, \quad (49)$$

where  $\langle \cdot \rangle_\rho$  represents the average over trajectories given by the Langevin equation

$$\frac{d}{dt} \rho = \alpha \rho - \beta \rho^2 + \sqrt{\frac{\alpha \rho + \beta \rho^2}{\Omega}} \cdot \eta \quad (50)$$

under the initial condition  $\rho(0) = \rho_i$ . This result coincides with the result obtained by any system-size expansion methods for the master equation. Since (49) holds for any  $\tau$ , the variable  $\rho$  defined by  $\mu/\Omega$  corresponds to the density in this description.

## 5. Effects of diffusion

In the main part of this paper, we have focused on the case where diffusion processes are sufficiently fast. This formulation can be extended to cases with spatially heterogeneous fluctuations. The simplest example in such systems is given by a one-dimensional lattice model, which we shall explain.

Let  $\Lambda = \{i | i = 0, 1, \dots, L\}$  be a one-dimensional lattice. The reaction  $X \rightleftharpoons X + X$  occurs on each site in the lattice, and a particle moves to a nearest-neighbor site at rate  $d$ . The state of the system is specified by a set of particle numbers on each site,  $\mathbf{n} = (n_1, \dots, n_L)$ . Boundary conditions are assumed appropriately, depending on the situation we consider. We then write the master equation for  $P(\mathbf{n}, t)$  and transform it to the equation for a single vector. The time-evolution operator  $\hat{H}$  in the equation for this vector is given by

$$\hat{H} = d \sum_{(ij) \in \mathcal{B}} (\hat{a}_i^\dagger - \hat{a}_j^\dagger)(\hat{a}_i - \hat{a}_j) + \sum_{i \in \Lambda} [-\alpha(\hat{a}_i^\dagger - 1)\hat{a}_i^\dagger \hat{a}_i - \beta(1 - \hat{a}_i^\dagger)\hat{a}_i^\dagger \hat{a}_i^2], \quad (51)$$

where  $\mathcal{B}$  represents a set of all the nearest-neighbor pairs, and  $\hat{a}_i^\dagger$  and  $\hat{a}_i$  are introduced on each site in a manner similar to  $\hat{a}^\dagger$  and  $\hat{a}$  in section 2.

Now, as a straightforward extension of the formulation discussed in section 4, we define a real variable  $\mu_i(t)$  which can be identified with the particle number on a site  $i$  at time  $t$ . Our central question for this model is to derive an *effective* stochastic system which is defined as the simplest description reproducing large-distance and long-time behavior. More explicitly, the effective stochastic system might be defined by a Langevin equation which provides a large deviation functional for the transition probability of a density field. Since the large deviation functional is related to an effective action in the field-theoretical language [20, 21], the question is equivalent to an identification of the fixed point in the renormalization group flow. However, as far as we know, a concrete calculation based on such a formulation has not yet been reported, and the large deviation functional in general cases cannot be obtained by conventional approaches used in [1, 3].

Putting aside developing such a theoretical framework, we here present a conventional derivation by considering a special situation. We first assume that one-site corresponds to a coarse-grained cell in which diffusion processes are sufficiently fast so that chemical components can be thought to be uniform in the cell. Let  $\Delta x$  be a dimensionless physical

size of the cell by setting a reaction length as unity (it thus corresponds to  $\Omega$  in the previous sections). As discussed in the first paragraph of section 2, we then replace  $\beta$  in (51) with  $\tilde{\beta}/\Delta x$  under the assumption  $\Delta x \gg 1$ . We also assume that there exists a density field  $\rho(x, t)$ ,  $0 \leq x \leq L\Delta x$ , that satisfies  $\rho(i\Delta x, t) = \mu_i(t)/(\Delta x)$  and  $\sup_x |\partial_x^k \rho(x, t)|(\Delta x)^k = O(\epsilon^k)$  with a small positive constant  $\epsilon$ . The latter condition means that the density field is slowly varying in space. Since the longest wavelength of fluctuations is  $O(L\Delta x)$ ,  $\epsilon$  corresponds to  $1/L$ . We thus consider the case  $L \gg 1$ . By repeating the argument in section 4 with the replacement of  $\Omega$  with  $\Delta x$ , one may derive

$$P(\mathbf{n}^{\text{fin}}, \tau | \mathbf{n}^{\text{init}}, 0) \simeq \left\langle \prod_{i=1}^L \delta \left( \rho(i\Delta x, \tau) - \frac{n_i^{\text{fin}}}{\Delta x} \right) \right\rangle_{\rho}, \quad (52)$$

where  $\simeq$  represents the equality valid asymptotically in the limit  $\Delta x \gg 1$ , and  $\langle \cdot \rangle_{\rho}$  represents the average over trajectories of a space-discretized form of the spatially extended Langevin equation

$$\frac{\partial \rho}{\partial t} = \alpha \rho - \tilde{\beta} \rho^2 + \tilde{d} \partial_x^2 \rho + \sqrt{\alpha \rho + \tilde{\beta} \rho^2} \cdot \xi_1 + \partial_x (\sqrt{2\tilde{d} \rho} \cdot \xi_2), \quad (53)$$

under the initial condition  $\rho(i\Delta x, 0) = n_i^{\text{init}}/\Delta x$ . Here, we defined  $\tilde{d} \equiv d(\Delta x)^2$  and  $\xi_1$  and  $\xi_2$  as noises satisfying

$$\langle \xi_i(x, t) \xi_j(x', t') \rangle = \delta_{i,j} \delta(x - x') \delta(t - t'). \quad (54)$$

We note that the discretized form of (54) is

$$\langle \xi_i(\ell \Delta x, t) \xi_j(\ell' \Delta x, t') \rangle = \delta_{i,j} \delta_{\ell,\ell'} \frac{1}{\Delta x} \delta(t - t'). \quad (55)$$

Although these arguments are standard, the validity of the derivation is restricted. Formally speaking, we focus on the limiting case  $L \rightarrow \infty$ ,  $d \rightarrow 0$ ,  $\beta \rightarrow 0$ ,  $d/\beta^2 \simeq \alpha \simeq O(1)$  for the lattice model (51). It is stimulating to analyze (51) together with a renormalization group idea so as to derive a hydrodynamic description for general cases.

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### Appendix. Path probability of a particle number

The probability of a particle-number trajectory  $\{n(t)\}$  is also expressed within the Doi–Peliti formalism. We briefly explain the expression in this appendix because there are no references mentioning the result explicitly. (See [15] for a related discussion.) We start with an expression for the probability  $P_n(t)$ :

$$P_{n_t}(\tau) = \lim_{\Delta t \rightarrow 0} \left( \prod_{t=\Delta t}^{\tau-\Delta t} \sum_{n_t=0}^{\infty} \right) \left( \prod_{t=0}^{\tau-\Delta t} K(n_t \rightarrow n_{t+\Delta t}) \right) P_{n_i}(t) \Big|_{n_0=n_i, n_{\tau}=n_t}, \quad (A.1)$$

where  $K(n \rightarrow m)$  is equivalent to the previously defined transition probability (35) applied for a small time step  $\Delta t$ :

$$K(n \rightarrow m) = \frac{1}{m!} \langle m | e^{-\hat{H} \Delta t} | n \rangle, \quad (A.2)$$

and we used the most elementary decomposition of unity  $\mathbf{1} = \sum_{n=0}^{\infty} |n\rangle\langle n|/n!$ . Rewriting  $K(n \rightarrow m)$  as

$$\begin{aligned} K(n \rightarrow m) &= \sum_{\sigma=-\infty}^{\infty} \frac{1}{(n+\sigma)!} \langle n+\sigma | e^{-\hat{H}\Delta t} | n \rangle \delta_{m,n+\sigma} \\ &= \int_{-\pi}^{\pi} \frac{d\theta}{2\pi} e^{-i\theta(m-n)} \sum_{\sigma=-\infty}^{\infty} \frac{1}{(n+\sigma)!} e^{i\theta\sigma} \langle n+\sigma | e^{-\hat{H}\Delta t} | n \rangle, \end{aligned}$$

we express the transition probability during a small time interval  $\Delta t$  as

$$K(n \rightarrow m) = \int_{-\pi}^{\pi} \frac{d\theta}{2\pi} \exp \left[ -i\theta(m-n) - \sum_{\sigma} L(i\theta, n; \sigma) \Delta t \right], \quad (\text{A.3})$$

where  $L(i\theta, n; \sigma)$  was defined as

$$L(i\theta, n; \sigma) = \frac{1}{(n+\sigma)!} \langle n+\sigma | \hat{H} | n \rangle e^{i\theta\sigma}. \quad (\text{A.4})$$

Expression (A.1) with (A.3) and (A.4) provides the probability of a trajectory  $\{n(t)\}$ . On the basis of this expression, one may develop a system-size expansion as we did in section 4. This formulation might be regarded as a sophisticated version of the discussion in [2].

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