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Fluctuations and correlations in a diffusion-reaction system: Unified description of internal fluctuations and external noise

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The one-dimensional single-species diffusion-limited-coagulation process with irreversible random particle input ($A \leftrightarrow A+A$ reversibly and $B \rightarrow A$ irreversibly), under the influence of external fluctuations in the system parameters, is formulated in terms of a closed and linear partial-differential equation. Our theoretical treatment includes both internal fluctuations and external noise simultaneously and without approximation, allowing investigation of the interplay of their effects on the macroscopic behavior of this diffusion-reaction system. For the reversible model with the *rate* of the $A \rightarrow A+A$ reaction fluctuating between two values as a Markov stochastic process, we solve the system exactly. We observe that spatially homogeneous macroscopic fluctuations in the system parameters can induce microscopic spatial correlations in the nonequilibrium steady state. Direct Monte Carlo simulations of the microscopic dynamics are presented, confirming the theoretical analysis and directly illustrating the external-noise-induced spatial correlations.

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I. INTRODUCTION

Fluctuations in macroscopic many-body systems arise from two sources. The discrete nature of the elementary constituents produces the so-called *internal* fluctuations, while random variations in the environment introduce *external* noise into the system. Theoretical treatment of reaction-diffusion systems often disregards both kinds of fluctuations. Internal fluctuations are neglected because they occur on microscopic length and time scales and are thus deemed unimportant for macroscopic phenomena (except for phase-transition phenomena), while external noise is usually excluded because its amplitude can be controlled in laboratory systems—it is usually considered more of a nuisance than an essential factor in the dynamics of the system. Results obtained during the last decade have demonstrated that internal fluctuations and external noise can each be a crucial factor in the qualitative macroscopic behavior of nonlinear interacting particle systems. External noise can postpone or advance instabilities, and may even give rise to transitions to states that cannot occur if the surroundings are free from random fluctuations [1,2]. Internal fluctuations can give rise to strong particle-particle correlations in transport-limited diffusion-reaction systems dominating the macroscopic dynamics [3–5].

Most theoretical studies that include effects of fluctuations consider either *only* internal fluctuations or *only* external noise, although a few authors have attempted to develop a unified description of both sources of stochastic behavior [6]. External noise is usually treated on a macroscopic level of description where mean-field rate equations are converted into stochastic differential equations by including random terms. For diffusion-reaction systems, inclusion of internal fluctuations requires a mesoscopic or microscopic description in terms of a random process for the particle numbers of the chemical species. These different levels of description lead to conceptual difficulties for a unified treatment of internal and external fluctuations, and so far no wholly satisfactory one exists.

In this paper we show that for a specific model system, namely the reversible coagulation-growth process ($A+A \leftrightarrow A$) with irreversible input ($B \rightarrow A$) in one spatial dimension, the above-mentioned difficulties can be overcome, and a unified description of internal fluctuations and external noise can be naturally formulated in terms of a quantity satisfying a *closed* kinetic equation without any approximation. We consider the coagulation reaction $A+A \rightarrow A$ in the *diffusion-controlled limit* where the particles coalesce immediately upon contact. These new results are generalizations of methods previously introduced for this diffusion-limited-reaction process [7,8], and we are able to obtain some exact results. Direct Monte Carlo simulation of the interacting particle system with externally imposed noise, also reported here, confirms our analysis quantitatively. We observe external-noise-induced spatial correlations in the steady-state microscopic particle positions, even in the case of spatially homogeneous external noise with no intrinsic spatial length scale: Environmental fluctuations can introduce new microscopic length scales into the system. This model provides an example of far-from-equilibrium constraints resulting in (partial) self-organization of a nonequilibrium steady state.

The opposite limit of the reversible process $A + A \leftrightarrow A$, the *reaction-controlled limit,* has been well studied in the past. The system can be satisfactorily described by a mean-field equation for the time-dependent concentration $c(t)$ if interparticle correlations remain small. If the macroscopic concentration is constant throughout space, then the kinetics is described in mean-field theory by the Verhulst equation [9]

$$\frac{dc}{dt}=r_1 c - r_2 c^2 , \tag{1.1}$$

where r_1 and r_2 are, respectively, the rate coefficients of the $A \rightarrow A + A$ and the $A + A \rightarrow A$ reactions. If the concentration also varies in space, then the mean-field dynamics is described by the Fisher reaction-diffusion equation [10]

$$\frac{\partial c}{\partial t}=D\nabla^2 c + r_1 c - r_2 c^2 , \tag{1.2}$$

with macroscopic diffusion coefficient D. The effect of either internal fluctuations or external noise on $A + A \leftrightarrow A$ in the reaction-controlled limit have been studied in Refs. [1] and [11].

The outline of the rest of this paper is as follows. In Sec. II we formulate the kinetic equation that provides a unified description of external noise and internal fluctuations for the one-dimensional coagulation process. We specialize to dichotomous (two-level) external noise in the birth rate for the reversible process in Sec. III, and derive a general result for the concentration in the stationary state. Section IV is devoted to the simplifying case that the birth rate fluctuates between zero and a positive value, and we determine the stationary state analytically, showing that external noise drives the system out of equilibrium. We also present the results of Monte Carlo simulations in this section to confirm and explicitly illustrate the phenomenon of external-noise-induced spatial correlations in this system. In Sec. V we study the system in the Poisson-white-noise limit, and Sec. VI contains a brief summary of the results, some observations, and a discussion of open problems.

II. MICROSCOPIC FORMULATION WITH EXTERNAL NOISE

We study a spatially one-dimensional system of diffusing particles that undergo the reversible coagulation-growth process $2A \leftrightarrow A$, and we consider the coagulation step in the diffusion-controlled limit, where the transport of particles constitutes the rate-limiting step. Furthermore, there may be an irreversible random input of particles, denoted schematically $B \rightarrow A$. Both of these processes are taken to be statistically spatially homogeneous in the system. This implies in particular that the diffusion coefficient D, the birth rate v, and the rate of particle input R are uniform in space.

The usual approach to interacting particle systems in statistical physics is to describe the system with a set of joint probability-density functions. No finite subset of this hierarchy is sufficient to fully characterize the system because of the usual closure problem: The temporal evolution of the one-particle density, or concentration, depends on the two-particle density, which in turn depends on higher particle densities. However, the closure problem can be circumvented for the model developed here. The key to an exact, closed, microscopic description is to abandon the particle densities and adopt an alternate quantity to characterize the system, namely the probability that a randomly chosen interval of length x is empty at time t, $E(x,t)$. For a fixed diffusion coefficient, birth rate, and input rate, the empty-interval probability obeys a linear partial-differential equation:

$$\begin{aligned}\partial_t E(x,t)&=2D\partial_{xx}E(x,t)+v\partial_x E(x,t)-RxE(x,t)\\ &\equiv L(D,v,R)E(x,t) ,\end{aligned} \tag{2.1}$$

with boundary conditions

$$E(0,t)=1 \text{ and } E(\infty,t)=0 . \tag{2.2}$$

The probability that a small interval of length dx is occupied is equal to $1-E(dx,t)$, and hence the concentration, or density, of particles is defined by

$$\rho(t)=-\left.\frac{\partial E(x,t)}{\partial x}\right|_{x=0} . \tag{2.3}$$

For a complete derivation from the microscopic definition of the processes, see Refs. [7] or [8]. We remark that this closed formulation is only valid for the diffusion-limited-coagulation process in one spatial dimension. We do not presently know how to apply this approach to obtain a closed formulation in higher dimensions or a closed formulation away from the diffusion-controlled limit.

Note that the concentration $\rho(t)$ is an ensemble average. It does not itself fluctuate, but it fully takes into account all the microscopic fluctuations in the system and any correlations that may develop. (We are considering an infinite system and have taken the thermodynamic limit.) Correlations can be characterized by the two-particle-correlation function, or alternatively by the interparticle distribution function (IPDF), $p(x,t)$, the probability density for finding the nearest particle a distance x from a given particle. The IPDF can be derived from the central quantity of our approach, i.e., the empty-interval probability $E(x,t)$:

$$\rho(t)p(x,t)=\frac{\partial^2 E(x,t)}{\partial x^2} . \tag{2.4}$$

All internal fluctuations are fully accounted for in the closed, exact microscopic kinetic Eq. (2.1). External noise can affect the system through random variations in the diffusion coefficient D, the birth rate v, or the input rate R. Typically the mechanisms of external noise are different for the various parameters, and it is therefore reasonable to assume that they are independent stochastic processes. We denote the external-noise processes by D_t, v_t, and R_t. Call the probability densities of these stochastic processes $p_D(D,t)$, $p_v(v,t)$, and $p_R(R,t)$ [if the fluctuations take on only discrete values, then $p_D(t)$,

$p_v(t)$, and $p_R(t)$ are probability vectors] and write their kinetic equations as:

$$\partial_t p_D = W_D p_D \ , \tag{2.5}$$

$$\partial_t p_v = W_v p_v \ , \tag{2.6}$$

and

$$\partial_t p_R = W_R p_R \ , \tag{2.7}$$

where W_D, W_v, and W_R are the evolution operators of the stochastic processes. An exact, closed, microscopic description of the reaction-diffusion system that takes into account its internal fluctuations as well as the external noise introduced by the surroundings can be formulated in terms of a natural extension of the central quantity of our approach, namely the *joint probability* $E(x,D,v,R,t)$, defined by

$$E(x,D,v,R,t)dD\,dv\,dR = \text{prob}[\text{interval of length } x \text{ is empty at time } t \text{ and } D_t \in (D,D+dD), \text{ and } v_t \in (v,v+dv), \text{ and } R_t \in (R,R+dR)] \ . \tag{2.8}$$

This joint probability obeys the kinetic equation

$$\partial_t E(x,D,v,R,t) = [L(D,v,R) + W_D + W_v + W_R]E(x,v,R,t) \ , \tag{2.9}$$

for statistically independent external fluctuations in the diffusion coefficient, the birth rate, and the rate of particle input. The boundary conditions are

$$E(0,D,v,R,t) = p_D(D,t)p_v(v,t)p_R(R,t) \tag{2.10}$$

and

$$E(\infty,D,v,R,t) = 0 \ . \tag{2.11}$$

The boundary conditions in the variables D, v, and R are specified once the stochastic processes D_t, v_t, and R_t are explicitly defined.

The (unconditional) empty-interval probability $E(x,t)$ is the marginal probability, given by

$$E(x,t) = \int dD \int dv \int dR\ E(x,D,v,R,t) \ . \tag{2.12}$$

(If the external-noise processes are discrete, the integration is replaced by a sum.) The ensemble-averaged concentration and IPDF's are recovered from the marginal empty-interval probability as in Eqs. (2.3) and (2.4).

We also have access to, for example, the conditional concentration of particles $\rho(t|D,v,R)$, given that the fluctuating parameters take on some specified values:

$$\rho(t|D,v,R) = -\frac{1}{E(0,D,v,R,t)} \left.\frac{\partial E(x,D,v,R,t)}{\partial x}\right|_{x=0} \ . \tag{2.13}$$

Similarly, the conditional IPDF at given parameter values, $p(x,t|D,v,R)$ is derived from

$$\rho(t|D,v,R)p(x,t|D,v,R) = \frac{1}{E(0,D,v,R,t)} \times \frac{\partial^2 E(x,D,v,R,t)}{\partial x^2} \ . \tag{2.14}$$

III. DICHOTOMOUS EXTERNAL NOISE

For the sake of concreteness we will restrict our attention to the case that the birth rate fluctuates like a dichotomous Markov process, also known as a random telegraph signal. A dichotomous noise takes on only two values, $v_t \in \{v_-, v_+\}$, and is Markovian if the probability of a jump from one state to the other state is independent of the time already spent in the state. This implies that the lifetime of each state is exponentially distributed. Let α and β be the jump frequencies. The kinetic equation then reads

$$\frac{d}{dt}\begin{bmatrix} P_- \\ P_+ \end{bmatrix} = \begin{bmatrix} -\beta & \alpha \\ \beta & -\alpha \end{bmatrix} \begin{bmatrix} P_- \\ P_+ \end{bmatrix} , \tag{3.1}$$

where $P_- = \text{prob}(v_t = v_-)$ and $P_+ = \text{prob}(v_t = v_+)$. The stationary probability for the dichotomous noise is given by

$$P_{s_+} = \frac{\beta}{\gamma}, \quad P_{s_-} = \frac{\alpha}{\gamma} \ , \tag{3.2}$$

with $\gamma = \alpha + \beta$. We illustrate the process in Fig. 1.

From now on we will assume that the external noise is a stationary random process. Its mean value is

$$\langle v_t \rangle = \frac{\alpha v_- + \beta v_+}{\gamma} \equiv v \ , \tag{3.3}$$

and its correlation function is

$$\langle v_t v_s \rangle - \langle v_t \rangle \langle v_s \rangle = \frac{\alpha\beta}{\gamma^2}(v_+ - v_-)^2 e^{-\gamma|t-s|} \ , \tag{3.4}$$

so the correlation time of the dichotomous noise is

$$\tau_{\text{cor}} = 1/\gamma \ . \tag{3.5}$$

The kinetic equation for the reaction-diffusion system subject to dichotomous noise in the birth rate, Eq. (2.9), takes the form

$$\frac{\partial}{\partial t}\begin{bmatrix} E_-(x,t) \\ E_+(x,t) \end{bmatrix} = \begin{bmatrix} L_- - \beta & \alpha \\ \beta & L_+ - \alpha \end{bmatrix} \begin{bmatrix} E_-(x,t) \\ E_+(x,t) \end{bmatrix} , \tag{3.6}$$

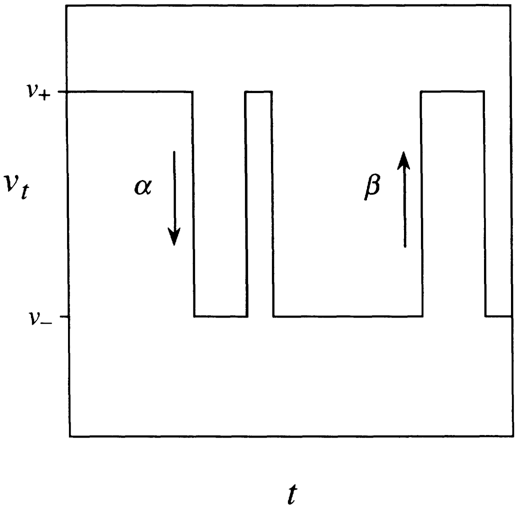


FIG. 1. Illustration of the dichotomous Markov process v_t. The process hops down from the upper level v_+ to the lower level v_- at rate α, and it hops up at rate β.

where

$$L_\pm = 2D\partial_{xx} + v_\pm \partial_x - Rx \ . \tag{3.7}$$

We may write these equations in dimensionless form by introducing rescaled space and time variables:

$$\begin{aligned} x' &= \frac{v}{2D}x, \quad t' = \frac{v^2}{2D}t \ , \\ v'_- &= \frac{v_-}{v}, \quad v'_+ = \frac{v_+}{v}, \quad R' = \frac{4D^2}{v^3}R \ , \\ \beta' &= \frac{2D}{v^2}\beta, \quad \alpha' = \frac{2D}{v^2}\alpha \ . \end{aligned} \tag{3.8}$$

The kinetic equations become (dropping the primes),

$$\partial_t E_- = \partial_{xx}E_- + v_-\partial_x E_- - RxE_- - \beta E_- + \alpha E_+ \tag{3.9}$$

and

$$\partial_t E_+ = \partial_{xx}E_+ + v_+\partial_x E_+ - RxE_+ + \beta E_- - \alpha E_+ \ . \tag{3.10}$$

The boundary conditions for Eqs. (3.9) and (3.10) are

$$E_-(0,t) = P_{s-} = \frac{\alpha}{\gamma}, \quad E_-(\infty,t) = 0 \tag{3.11}$$

and

$$E_+(0,t) = P_{s+} = \frac{\beta}{\gamma}, \quad E_+(\infty,t) = 0 \ . \tag{3.12}$$

Note that in dimensionless variables the ensemble-averaged concentration is

$$\rho'(t') = \frac{2D}{v}\rho\left(\frac{2Dt'}{v^2}\right) \tag{3.13a}$$

and is computed (again, dropping the primes)

$$\rho(t) = -\left.\frac{\partial E}{\partial x}\right|_{x=0} , \tag{3.13b}$$

where the unconditional empty-interval probability $E(x,t)$ is given by

$$E(x,t) = E_-(x,t) + E_+(x,t) \ . \tag{3.14}$$

We may further reduce the system of two coupled equations, Eqs. (3.9) and (3.10), to one evolution equation by eliminating E_+. Rewriting Eq. (3.9) as

$$\alpha E_+ = \partial_t E_- - \partial_{xx}E_- - v_-\partial_x E_- + RxE_- + \beta E_- \ , \tag{3.15}$$

and injecting this expression into Eq. (3.10), we obtain

$$\begin{aligned} &\partial_{tt}E_- + \gamma\partial_t E_- - 2\partial_{xxt}E_- - (v_+ + v_-)\partial_{xt}E_- + 2Rx\partial_t E_- \\ &\quad = -\partial_{xxxx}E_- - (v_+ + v_-)\partial_{xxx}E_- + (\gamma - v_+v_-)\partial_{xx}E_- + (\beta v_+ + \alpha v_-)\partial_x E_- \\ &\qquad + R\{2x\partial_{xx}E_- + [2 + (v_+ + v_-)x]\partial_x E_- + [v_+ - Rx^2 - \gamma x]E_-\} \ . \end{aligned} \tag{3.16}$$

Recall that the mean value of the birth rate is

$$\langle v_t \rangle \equiv v = \frac{\alpha v_- + \beta v_+}{\gamma} \ ,$$

which equals one with the scaling in Eq. (3.8). The variance is given by

$$w^2 \equiv \langle v_t^2 \rangle - v^2 = \frac{\alpha\beta}{\gamma^2}(v_+ - v_-)^2 \ , \tag{3.17}$$

so we can express the two values v_- and v_+ in terms of α, β, and w:

$$v_- = 1 - w\left[\frac{\beta}{\alpha}\right]^{1/2} , \tag{3.18}$$

$$v_+ = 1 + w\left[\frac{\alpha}{\beta}\right]^{1/2} . \tag{3.19}$$

Defining $\eta = \beta/\gamma$ and $\mu = 1 - 2\eta/\sqrt{\eta(1-\eta)}$, we obtain

$$v_+ + v_- = 2 + \mu w \ , \tag{3.20}$$

$$v_+v_- = 1 + \mu w - w^2 \ . \tag{3.21}$$

The equivalent form for Eq. (3.16) is then

$$\begin{aligned}
&\partial_{tt}E_- + \gamma\partial_t E_- - 2\partial_{xxt}E_- - (2+\mu w)\partial_{xt}E_- + 2Rx\partial_t E_- \\
&\quad = -\partial_{xxxx}E_- - (2+\mu w)\partial_{xxx}E_- + (\gamma - 1 - \mu w + w^2)\partial_{xx}E_- + \gamma\partial_x E_- \\
&\qquad + R\{2x\partial_{xx}E_- + [2+(2+\mu w)x]\partial_x E_- + [1+\sqrt{\eta^{-1}-1}w - Rx^2 - \gamma x]E_-\} \ .
\end{aligned} \tag{3.22}$$

We remark here that this linear partial-differential equation constitutes an exact formulation of the diffusion-reaction process under the influence of dichotomous Markov fluctuations in the birth rate. It is worth noting the simplicity to which this ostensibly nonlinear, stochastic, many-body problem has been reduced.

Now we focus on the steady state of the system ($\partial_t = 0$) and consider only the reversible model $2A \leftrightarrow A$ (i.e., $R=0$). The stationary form of Eq. (3.16) without particle input reads

$$\begin{aligned}
-\partial_{xxxx}E_- - (v_+ + v_-)\partial_{xxx}E_- + (\gamma - v_+ v_-)\partial_{xx}E_- \\
+ (\beta v_+ + \alpha v_-)\partial_x E_- = 0 \ .
\end{aligned} \tag{3.23}$$

With the usual ansatz for linear differential equations with constant coefficients

$$E_-(x) \propto e^{-kx} \ , \tag{3.24}$$

we obtain the characteristic polynomial for k,

$$-k^4 + (v_+ + v_-)k^3 + (\gamma - v_+ v_-)k^2 - (\beta v_+ + \alpha v_-)k = 0 \ . \tag{3.25}$$

In addition to the $k=0$ eigenvalue corresponding to an empty system, the eigenvalues are the roots of the cubic polynomial

$$k^3 - (v_+ + v_-)k^2 - (\gamma - v_+ v_-)k + (\beta v_+ + \alpha v_-) = 0 \tag{3.26}$$

Only positive roots or roots with a positive real part are acceptable in light of the boundary condition as $x \to \infty$ in Eq. (3.11). The sign structure of Eq. (3.26) is of the form $+,-,\pm,+$, so the number of sign changes is always equal to two and Descartes's rule implies that there are either two or zero real positive roots. Although general cubic equations can be solved exactly, the complicated explicit form of the roots is not very enlightening for this example, and we therefore specialize to a simplifying case in the next section.

We end this section with the proof of a (somewhat surprising) general result for this model: External dichotomous Markov fluctuations of the birth rate in the reversible model do not affect the stationary average value of the concentration, provided that both the jump frequencies are nonvanishing and the variance is finite. To show this we begin with an alternative formulation of the kinetic equations in terms of the total empty-interval probability $E = E_+ + E_-$ and the difference quantity

$$Q \equiv \alpha E_+ - \beta E_- \ . \tag{3.27}$$

Adding Eqs. (3.9) and (3.10), and noting that

$$E_- = \frac{1}{\gamma}(\alpha E - Q) \tag{3.28}$$

and

$$E_+ = \frac{1}{\gamma}(\beta E + Q) \ , \tag{3.29}$$

we obtain the evolution equation for the total empty-interval probability:

$$\partial_t E = \partial_{xx}E + \partial_x E + \frac{1}{\gamma}(v_+ - v_-)\partial_x Q - RxE \ . \tag{3.30}$$

Multiplying Eq. (3.9) by $-\beta$ and adding it to Eq. (3.10) multiplied by α, we find

$$\begin{aligned}
\partial_t Q = \partial_{xx}Q + \frac{\alpha\beta}{\gamma}(v_+ - v_-)\partial_x E + \frac{1}{\gamma}(\beta v_- + \alpha v_+)\partial_x Q \\
- RxQ - \gamma Q \ .
\end{aligned} \tag{3.31}$$

(Note that the coefficient of the $\partial_x Q$ term is *not* equal to $\langle v_t \rangle$.) Let us again focus on the reversible case, $R=0$. From Eq. (3.30) we obtain

$$\partial_x Q = \frac{\gamma}{v_+ - v_-}(\partial_t E - \partial_{xx}E - \partial_x E) \ . \tag{3.32}$$

Differentiating Eq. (3.31), with $R=0$, with respect to x and using Eq. (3.32) to eliminate $\partial_x Q$, we obtain a closed equation for E:

$$\begin{aligned}
&\partial_{tt}E + \gamma\partial_t E - 2\partial_{xxt}E - (v_+ + v_-)\partial_{xt}E \\
&\quad = -\partial_{xxxx}E - (v_+ + v_-)\partial_{xxx}E \\
&\qquad + (\gamma - v_+ v_-)\partial_{xx}E + (\beta v_+ + \alpha v_-)\partial_x E \ ,
\end{aligned} \tag{3.33}$$

which coincides, as expected, exactly in structure with Eq. (3.16) for $R=0$.

The main interest in using this formulation in terms of E and Q lies in the following observation. Focusing again on the stationary case, we obtain

$$\begin{aligned}
\partial_x Q &= \frac{\gamma}{v_+ - v_-}(-\partial_{xx}E - \partial_x E) \\
&= \frac{\sqrt{\alpha\beta}}{w}(-\partial_{xx}E - \partial_x E) \ .
\end{aligned} \tag{3.34}$$

This equation is easily integrated yielding

$$Q(x) = C - \frac{\sqrt{\alpha\beta}}{w}[E'(x) + E(x)] \ . \tag{3.35}$$

The boundary conditions in Eqs. (3.11) and (3.12) imply that

$$Q(0)=\alpha E_+(0)-\beta E_-(0)=\alpha\frac{\beta}{\gamma}-\beta\frac{\alpha}{\gamma}=0 . \tag{3.36}$$

Therefore we obtain, from Eq. (3.35), that

$$C=\frac{\sqrt{\alpha\beta}}{w}[\partial_x E(x)|_{x=0}+E(0)] . \tag{3.37}$$

Using the boundary condition for the empty-interval probability in Eq. (2.2) and the expression in Eq. (3.13) for the concentration, we rewrite Eq. (3.37) in the form

$$C=\frac{\sqrt{\alpha\beta}}{w}(-\rho_s+1) . \tag{3.38}$$

(Here ρ_s is the dimensionless stationary concentration.) The boundary conditions in Eqs. (3.12) and (3.13) imply also that

$$Q(\infty)=\alpha E_+(\infty)-\beta E_-(\infty)=0 . \tag{3.39}$$

Furthermore, $E(\infty)=0$ and $\partial_x E\rightarrow 0$ as $x\rightarrow\infty$. Hence we conclude that

$$0=Q(\infty)=C . \tag{3.40}$$

We deduce that

$$\rho_s=1 \tag{3.41}$$

as long as $\sqrt{\alpha\beta}/w\neq 0$. In dimensioned variables this is

$$\rho_s=\frac{\langle v\rangle}{2D} . \tag{3.42}$$

Away from the limiting (somewhat "degenerate") cases where $\alpha=0$, $\beta=0$, or $w=\infty$, the stationary density equals the *equilibrium* value for a system with the same *average* birth rate. While external dichotomous noise in the birth rate does not thus modify the average steady-state density, it does affect the functional form of the stationary empty-interval probability $E(x)$ and hence the stationary interparticle distribution function $p(x)$. That is, the slope of the empty-interval probability at $x=0$ is independent of the noise characteristics, but the behavior of $E(x)$ for $x\neq 0$ depends on the noise amplitude and jump frequencies: The eigenvalues k [the roots of the cubic polynomial in Eq. (3.26)] vary with the values of the noise parameters. Since the IPDF is proportional to the second derivative of $E(x)$, it too will deviate from its equilibrium form in the presence of fluctuations in the birth rate. As mentioned earlier, the general solution of the cubic equation for the k's is not very revealing, so we consider a special case in the next section.

IV. COAGULATION WITH RANDOMLY VANISHING BIRTHRATE

The birth rate v must be a non-negative quantity, which implies that $v_-\geq 0$. In this section we consider the extreme but simplifying case that the birth rate fluctuates between zero $v_-=0$) and a positive value ($v_+>0$). The birth process is then switching on and off randomly in time.

The scaling in Eq. (3.9) has the consequence that the dimensionless birth rate always has mean value one, $\langle v_t\rangle=v=1$. With $v_-=0$, this implies that

$$v_+=\frac{1}{\eta}=\frac{\gamma}{\beta} \tag{4.1}$$

and

$$w=\frac{\sqrt{\alpha\beta}}{\gamma}v_+=\left[\frac{\alpha}{\beta}\right]^{1/2}=\left[\frac{1}{\eta}-1\right]^{1/2} . \tag{4.2}$$

Equation (3.26) for the eigenvalues then reduces to

$$k^3-\frac{\gamma}{\beta}k^2-\gamma k+\gamma=0 , \tag{4.3}$$

which we write as

$$\Psi(k)\equiv k^3-\frac{\gamma}{\beta}k^2-\gamma k=-\gamma . \tag{4.4}$$

Defining $H\equiv\eta^{-1}$, we have

$$\Psi(k)\equiv k^3-Hk^2-\gamma k=-\gamma . \tag{4.5}$$

Because $\Psi'(0)=-\gamma$, the cubic function Ψ has a minimum for positive values of k and $\Psi_{\min}<0$. In the Appendix we show that $\Psi_{\min}\leq-\gamma H$, implying that for all values of the noise parameters the characteristic polynomial in Eq. (4.3), or in Eq. (4.5), has two positive roots. (Recall that $H=\eta^{-1}=\gamma/\beta>1$, so $-\gamma H<-\gamma$. The borderline case $\gamma=\beta$ is unphysical because then the mean life-time of the state v_+ is infinite.)

Let k_1 and k_2 denote the two positive roots of Eq. (4.3). The steady-state expression for E_- is given by

$$E_-(x)=C_1 e^{-k_1 x}+C_2 e^{-k_2 x} . \tag{4.6}$$

From Eq. (3.16) we obtain the corresponding expression for E_+,

$$\alpha E_+=-\partial_{xx}E_-+\beta E_- \tag{4.7}$$

or

$$\alpha E_+(x)=C_1(\beta-k_1^2)e^{-k_1 x}+C_2(\beta-k_2^2)e^{-k_2 x} . \tag{4.8}$$

The constants C_1 and C_2 are determined by the boundary conditions at $x=0$ in Eqs. (3.12) and (3.13):

$$E_-(0)=P_{s-}=\frac{\alpha}{\gamma}$$

and

$$E_+(0)=P_{s+}=\frac{\beta}{\gamma} .$$

From Eqs. (4.6) and (4.8) we obtain

$$C_1+C_2=\frac{\alpha}{\gamma} \tag{4.9}$$

and

$$C_1(\beta-k_1^2)+C_2(\beta-k_2^2)=\frac{\alpha\beta}{\gamma} , \tag{4.10}$$

which are equivalent to

$$C_1+C_2=\frac{\alpha}{\gamma} \tag{4.11}$$

and

$$C_1k_1^2+C_2k_2^2=0\ . \tag{4.12}$$

The solution of Eqs. (4.11) and (4.12) is

$$\begin{bmatrix} C_1 \\ C_2 \end{bmatrix} = \frac{\alpha}{\gamma}\frac{1}{k_2^2-k_1^2}\begin{bmatrix} k_2^2 \\ -k_1^2 \end{bmatrix} \tag{4.13}$$

The stationary empty-interval probability,

$$E(x)=\frac{1}{\gamma}\frac{1}{k_2^2-k_1^2}[k_2^2(\gamma-k_1^2)e^{-k_1x} - k_1^2(\gamma-k_2^2)e^{-k_2x}]\ , \tag{4.14}$$

and the stationary IPDF,

$$p(x)=\frac{1}{\rho}\frac{\partial^2E(x)}{\partial x^2} = \frac{1}{\gamma}\frac{k_1^2k_2^2}{k_2^2-k_1^2}[(\gamma-k_1^2)e^{-k_1x}-(\gamma-k_2^2)e^{-k_2x}]\ , \tag{4.15}$$

are both sums of two monotone decaying exponentials. The first derivative of the empty-interval probability is

$$\frac{\partial E(x)}{\partial x}=\frac{1}{\gamma}\frac{-k_1k_2}{k_2^2-k_1^2}[k_2(\gamma-k_1^2)e^{-k_1x} - k_1(\gamma-k_2^2)e^{-k_2x}]\ , \tag{4.16}$$

and using Eq. (4.3) we can confirm explicitly that, indeed, $E'(0)=-1$.

These results provide a specific realization of our remarks at the end of Sec. III: Although the stationary density is independent of the noise in the birth rate $[\rho=-E'(0)=1]$, $E(x)$ and $p(x)$ deviate from their equilibrium forms [12] (also see Ref. [4]) for fixed birth rate:

$$E^{\text{equil}}(x)=e^{-x}\ , \tag{4.17}$$

and

$$p^{\text{equil}}(x)=e^{-x}\ . \tag{4.18}$$

Note in particular that the external noise drives the IPDF away from the simple exponential distribution, corresponding to a totally random Poisson distribution of particles on the line, which maximizes the entropy. A nonexponential IPDF indicates microscopic spatial correlations in the particle positions, correlations which are absent in the true equilibrium state. External noise destroys the property of detailed balance and induces a stationary *nonequilibrium* state in this diffusion-reaction process.

In Fig. 2 we plot $p(x)$ versus x for the case $\gamma=10$ and $\beta=1$ corresponding to a birth-rate process v_t which, on average, spends $\frac{1}{10}$ of the time at 10 times its average value. The equilibrium IPDF is plotted for comparison. The particle positions are strongly correlated in this nonequilibrium steady state. The nonequilibrium $p(x)$ is larger than $p^{\text{equil}}(x)$ for small x and large x, indicating that we would expect to see relatively more of those smaller and larger gaps between adjacent particles, as compared to a completely uncorrelated distribution of particles.

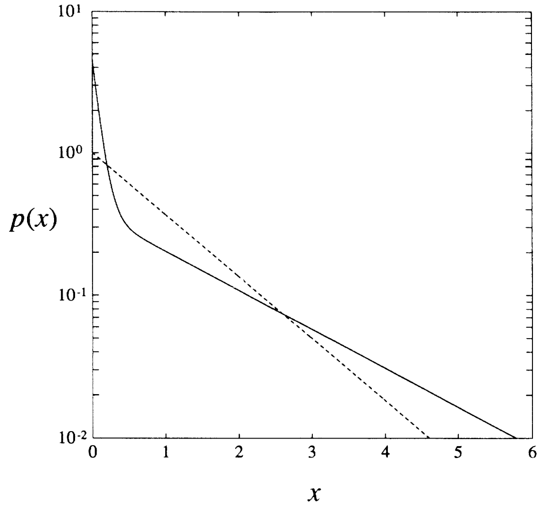


FIG. 2. Interparticle distribution functions for the noisy system (continuous curve) and an equilibrium system with the same concentration (dashed). The noise parameters are, in dimensionless units, $\gamma=10$ and $\beta=1$.

We have performed direct Monte Carlo simulations of the reversible diffusion-limited-coagulation process with the birth rate fluctuating as a dichotomous Markov process between $v_-=0$ and $v_+>0$. The simulation algorithm is the same as that in Ref. [5], with the straightforward addition of the fluctuating birth rate. From a practical point of view the microscopic simulation of the system with macroscopic noise required significantly more computer time than, say, a simulation of the relaxation

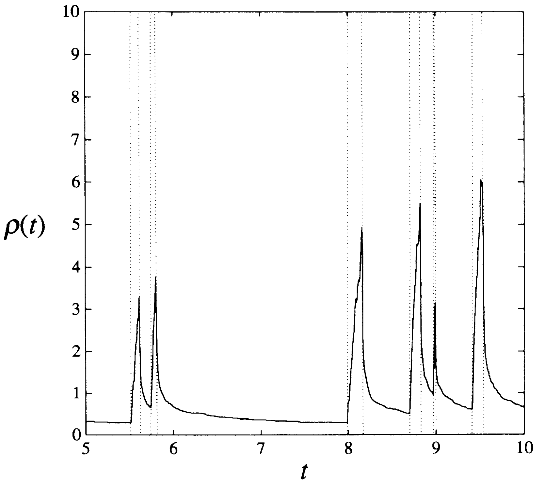


FIG. 3. Concentration vs time from a Monte Carlo simulation. The dotted vertical lines indicate where the birth rate switches between $v_-=0$ and $v_+=10$. The noise parameters are, in dimensionless units, $\gamma=10$ and $\beta=1$.

from one equilibrium state to another. This is because we consider external fluctuations on a "macroscopic" time scale that is not fast in comparison with the relevant microscopic relaxation times. The simulation must run long enough on this macroscopic time scale for the external noise process to reach its steady state, which may be many microscopic relaxation times.

In Fig. 3 we plot the concentration versus time for the reversible diffusion-limited-coagulation process simulated on a spatial lattice of 10^5 sites. The vertical (dotted) lines indicate the times where the birth rate hopped between v_+ and $v_-=0$. As expected, the time-dependent fluctuating concentration follows the birth rate fluctuations. The parameter values are the same as those used in Fig. 2. That is, in our dimensionless units v_t jumps up from 0 to $v_+=10$ at rate 1, and down from v_+ to 0 at rate 9. The exponential relaxation rate of an equilibrium system at the maximum birth rate ($v_+=10$) is 100 in these units, so the noise may be considered relatively slow on this scale. Although the average concentration of particles is 1 in our dimensionless units, in units of the inverse lattice spacing the average concentration is 10^{-3} and the equilibrium concentration at the maximum birth rate would be 10^{-2}. Thus comparison with our spatially continuous theory is appropriate. The Monte Carlo data for the average IPDF is plotted as a histogram in Fig. 4, along with the theoretical curve [13]. The agreement fully confirms our analysis.

It is also interesting to ask if the spatial correlations in the nonequilibrium steady state manifest themselves in a way which is visually apparent, without recourse to any statistical analysis (such as binning into probability distributions). In order to investigate this question we have looked at a number of "snapshots" of the particle configurations during the simulation.

One such typical snapshot is presented in Fig. 5(a), where the horizontal dimension is the spatial position and a vertical line represents the presence of a particle. This snapshot was taken while the birth process happened to be turned off ($v_t=v_-=0$), and there are 60 particles shown, distributed according to the system's own self-organization scheme. The spatial distribution in Fig. 5(a) should be qualitatively compared to that in Fig. 5(b), where we have distributed particles randomly on the same length of line at the same concentration. The random distribution is that which would be realized in an *equilibrium* system at the same density. The snapshot from the nonequilibrium steady state clearly shows a more even spacing of particles than the equilibrium distribution.

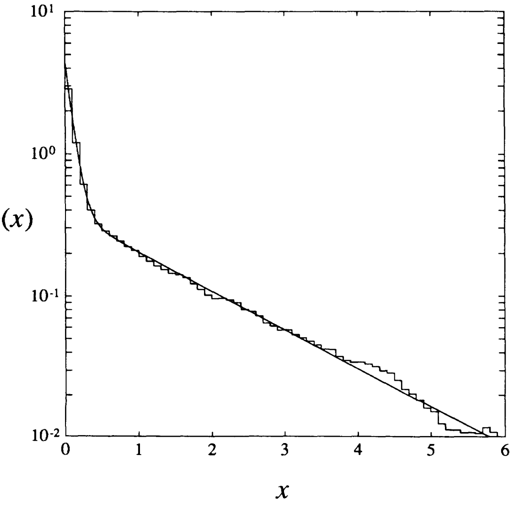
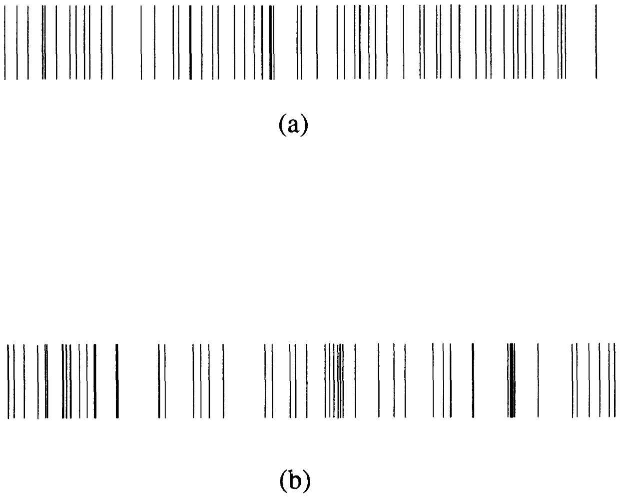


FIG. 4. Interparticle distribution function, theory (continuous curve), and Monte Carlo data (histogram). The system parameters are, in dimensionless units, $\gamma=10$ and $\beta=1$.


(a)

(b)

FIG. 5. Snapshots of the spatial distribution of particles. (a) The nonequilibrium system subject to noise in the case that the birth rate is in the off state. (b) A totally random distribution of particles at the same concentration, as is realized in an equilibrium state. The particles are seen to be more evenly spaced in the nonequilibrium state relative to the equilibrium state. The noise parameters are, in dimensionless units, $\gamma=10$ and $\beta=1$.

This self-imposed isolation of the particles is reflected in the IPDF conditioned on the event that $v_t=v_-$, i.e., the conditional IPDF $p_-(x)$. Recalling Eqs. (2.13) and (2.14), and using Eqs. (4.6) and (4.13), we compute

$$p_-(x)=\frac{1}{\left.\frac{-\partial E_-}{\partial x}\right|_{x=0}}\frac{\partial^2 E_-(x)}{\partial x^2}$$

$$=\frac{k_1k_2}{k_2-k_1}(e^{-k_1x}-e^{-k_2x}) . \tag{4.19}$$

We plot this IPDF, for the parameter values used in the Monte Carlo simulation, along with that corresponding to an equilibrium distribution of particles at the same density in Fig. 6. The vanishing of the nonequilibrium IPDF as $x\rightarrow 0$ is the hallmark of this statistical repulsion.

It is also worthwhile to compare the conditional IPDF in Eq. (4.19) with the average one in Eq. (4.15) (plotted in Figs. 2 and 4). On average, it is *more* likely to find particles close together in the nonequilibrium state, in contrast to conclusions drawn from the snapshot in Fig. 5. This is because when the birth rate is turned on (which is

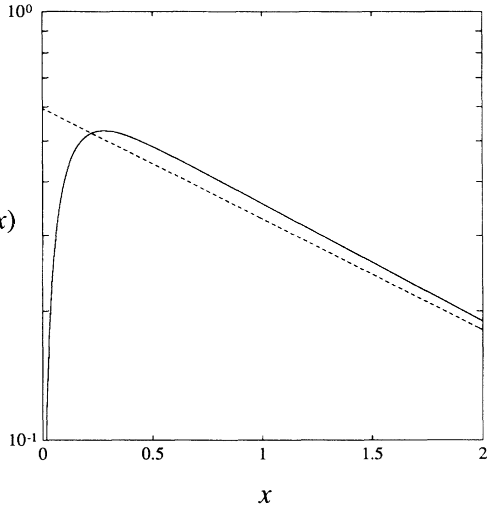


FIG. 6. Conditional IPDF, given that the birth rate v_t is in the off state (continuous curve). The IPDF for a random distribution of particles at the same density (dashed line) is shown for comparison. The noisy-system parameters are, in dimensionless units, $\gamma = 10$ and $\beta = 1$.

only one tenth of the time in this simulation), each of the relatively well separated particles in Fig. 5(a) spawns its own local "clump" of particles. The density of particles within the clumps is then much higher than the "clump density," i.e., the density of the well separated individual particles when the birth rate is zero. This effect is directly illustrated in the snapshot in Fig. 7(a), and in the conditional IPDF when the birth process is on is plotted in Fig. 8. The unconditional IPDF is just the appropriately weighted average of the two conditional distributions.

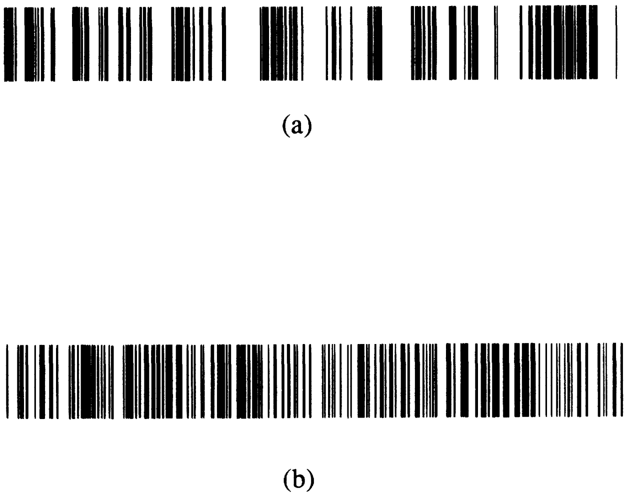


FIG. 7. Snapshots of the spatial distribution of particles. (a) The nonequilibrium system in the case that the birth rate is in the on state. (b) A completely random distribution of the same number of particles, as in an equilibrium state at the same density. The particles are seen to be more clumped together in the nonequilibrium system relative to the equilibrium distribution. The noisy-system parameters are, in dimensionless units, $\gamma = 10$ and $\beta = 1$.

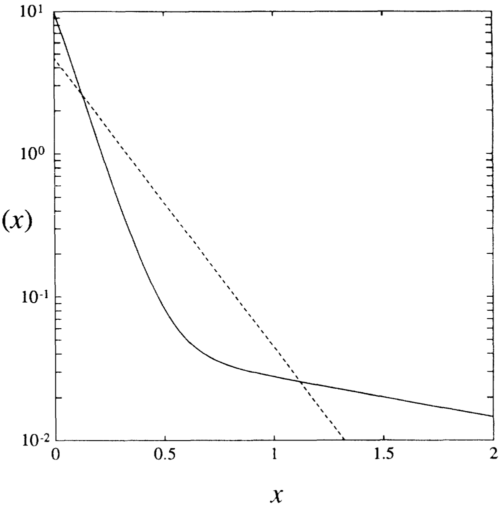


FIG. 8. Conditional IPDF, given that the birth rate v_t is in the on state (continuous curve). The IPDF for a random distribution of particles at the same density (dashed line) is shown for comparison. The noisy-system parameters are, in dimensionless units, $\gamma = 10$ and $\beta = 1$.

The clumping effect in the nonequilibrium system can be quantified more precisely in the extreme case of very short, but high-intensity bursts of the birth process. This is the Poisson-white-noise limit of the stochastic process v_t and is developed in the next section.

V. THE POISSON-WHITE-NOISE LIMIT

In applications the typical time scale of the noise, e.g., the correlation time, may be short compared to the intrinsic time scale of the system. Such situations are commonly modeled by white-noise processes where the limit of vanishing correlation time is taken $\tau_{\text{cor}} \to 0$. In our problem the familiar Gaussian white noise cannot be used because it is unbounded from below, and the birth rate v must remain non-negative. However, we may consider the Poisson-white-noise limit of the dichotomous Markov process [14]. In the limit

$$v_+ \to \infty, \quad \alpha \to \infty, \quad \text{such that } \frac{v_+}{\alpha} = \sigma = O(1) , \tag{5.1}$$

the dichotomous noise goes over into Poisson white noise with exponentially distributed weights. In this limiting situation the lower state v_- can be considered a baseline value of the birth rate, which is randomly interrupted, at frequency β, by bursts of high birth rate of value v_+. The lifetime of these bursts is exponentially distributed with mean α^{-1}. The weight of a burst is defined as the area of the pulse, which is exponentially distributed with mean v_+/α. Thus the Poisson-white-noise limit corresponds to increasing the amplitude of the burst and decreasing its average lifetime, such that the mean weight of the burst—or mean area of the pulse—remains constant. Poisson white noise can be pictured as a random sequence of δ spikes superimposed on the baseline. These spikes occur with frequency β and have an exponentially distributed weight of mean value σ. Because

$$1=v=\frac{\alpha v_- +\beta v_+}{\gamma}=\frac{\alpha v_- +\beta\alpha\sigma}{\gamma}=\frac{\alpha}{\gamma}(v_- +\beta\sigma)\ ,$$

and $\alpha/\gamma\to 1$ in the Poisson-white-noise limit, we find that the lower bound for the Poisson-white-noise process is

$$v_- = 1-\beta\sigma\ , \tag{5.2}$$

and we must impose

$$\beta\sigma\le 1 \tag{5.3}$$

in order to ensure the non-negativity of v_t.

As the Poisson-white-noise limit is approached we keep only the dominant terms in the coefficients of the characteristic polynomial in Eq. (3.26), which reduces to

$$k^3-\alpha[\sigma k^2+(1-\sigma+\beta\sigma^2)k-1]=0\ . \tag{5.4}$$

To solve this cubic equation, we make the following scaling ansatz:

$$k=\alpha^m\kappa\ . \tag{5.5}$$

Then

$$\alpha^{3m}\kappa^3-\alpha\sigma\alpha^{2m}\kappa^2-\alpha(1-\sigma+\beta\sigma^2)\alpha^m\kappa+\alpha=0\ , \tag{5.6}$$

and the powers of α are $3m$, $2m+1$, $m+1$, and 1. There are two and only two ways of balancing the powers of α:

$$\alpha^{3m}=\alpha^{2m+1}\Longrightarrow m=1 \tag{5.7}$$

or

$$\alpha^{2m+1}=\alpha^{m+1}\Longrightarrow m=0\ . \tag{5.8}$$

With $m=1$ we obtain from Eq. (5.4), to dominant order,

$$\kappa^3-\sigma\kappa^2=0\ . \tag{5.9}$$

Using $m=0$ we find, to dominant order,

$$\sigma k^2+(1-\sigma+\beta\sigma^2)k-1=0\ . \tag{5.10}$$

Therefore near the Poisson-white-noise limit the roots of the characteristic equation are

$$k_1=\alpha\sigma\ , \tag{5.11}$$

$$k_{2,3}=-\frac{1}{2}\left\{\left[\frac{1}{\sigma}-1+\beta\sigma\right]\pm\left[\left[\frac{1}{\sigma}-1+\beta\sigma\right]^2+\frac{4}{\sigma}\right]^{1/2}\right\}. \tag{5.12}$$

Clearly the roots k_2 and k_3 are of opposite sign. The total eigenvalue spectrum in the vicinity of the Poisson-white-noise limit is given by $-,0,+,\to+\infty$.

Let us again consider the extreme but simplifying case that $v_-=0$. Then we find from Eq. (5.2) that

$$\beta=1/\sigma\ , \tag{5.13}$$

and the acceptable eigenvalues are

$$k_1=\frac{\alpha}{\beta}\ , \tag{5.14}$$

$$k_2=\frac{1}{2}\beta(\sqrt{1+4/\beta}-1)\ . \tag{5.15}$$

Applying the Poisson-white-noise limit to Eqs. (4.14), (4.15), and (4.16) we obtain (being careful to perform the appropriate differentiations before taking the limit)

$$E_{\rm PWN}(x)=e^{-k_2x}\ , \tag{5.16}$$

$$\frac{d}{dx}E_{\rm PWN}(x)=-\frac{k_2^2}{\beta}H(x)-k_2e^{-k_2x}\ , \tag{5.17}$$

and

$$p_{\rm PWN}(x)=k_2^2[e^{-k_2x}+\frac{1}{\beta}\delta(x)]\ , \tag{5.18}$$

where $H(0)=1$ and $H(x)=0$ for $x\ne 0$, and $\delta(x)$ is the Dirac δ function.

The interparticle distribution function shows that Poisson white noise in the birth rate leads to the clumping phenomena illustrated and discussed in the previous section. Because of the δ function in $p_{\rm PWN}$, the most probable distance to a neighboring particle is, on average, zero. The relatively long tail of $p_{\rm PWN}$, compared to an equilibrium system with the same concentration, signifies that there are large empty intervals between the clumps. Indeed, while the average distance between nearest neighbors is

$$\langle x\rangle=\int_0^\infty xp_{\rm PWN}(x)dx=1\ . \tag{5.19}$$

The *conditional* average distance between neighbors given that the gap is nonzero, which can be interpreted as the average distance between clumps, is

$$\langle x|x\ne 0\rangle=\frac{\int_{0^+}^\infty xp_{\rm PWN}(x)dx}{\int_{0^+}^\infty p_{\rm PWN}(x)dx}=k_2^{-1}>1\ .$$

We conclude that (on average) there is a clump density k_2 (<1), each with average concentration k_2^{-1} (>1) particles per clump.

Recall that the mean weight of the spikes in the Poisson noise is $\sigma=1/\beta$. The expression for $p_{\rm PWN}$ in Eq. (5.18) reveals that rare, but strong bursts of particle birth, i.e., small β, lead to the strongest clumping tendency. As the bursts become more frequent, but less strong, the system becomes less self-ordered, and we recover the totally random *equilibrium* state as $\beta\to\infty$. In this limit the external-noise time scales have all become faster than any of the microscopic time scales, and the system simply responds to the *average* state of the environment as if there were no fluctuations.

VI. DISCUSSION

We have achieved a unified, closed description of internal fluctuations and external noise for an interesting model system, the reversible diffusion-limited-coagulation reaction $A+A\leftrightarrow A$ with irreversible input $B\to A$ in one spatial dimension. We have derived explicit results for the stationary state of the reversible process with external dichotomous noise in the birth rate, and in particular we have studied the limiting case that the birth rate fluctuates between zero and a fixed positive value. External noise drives the system out of equilibrium to a stationary nonequilibrium state, and the interparticle distribution function is then no longer a simple exponential corresponding to a totally random distribution of particles on the line, the maximum-entropy state characteristic of equilibria. We conclude that spatially homogeneous noise induces spatial correlations in the system in the form of clumping: The particles tend to bunch together on average, leaving relatively large empty intervals in between. For the type of noise studied, this effect occurs without a threshold, and the deviation from the equilibrium increases smoothly with the strength of the noise.

There remain several open problems. First, in the absence of external noise, the strictly reversible process $A+A\leftrightarrow A$ exhibits an interesting dynamic transition in its relaxation kinetics when switching between equilibria of different values of the system parameters (see both Refs. [7] and [11]). The effect of external noise on the dynamics can be analyzed by a simple extension of the ansatz in Eq. (3.24), namely that $E(x,t)\propto e^{-\lambda t}e^{-kx}$. Second, the effects of external spatial disorder, frozen in time, can be investigated using our recent extension of the formalism to spatially inhomogeneous situations, as developed in Ref. [6].

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APPENDIX

We show that $\Psi_{\min}\leq-\gamma H$. The extrema of Ψ are the roots of

$$\Psi'(k)=3k^2-2Hk-\gamma=0\ . \tag{A1}$$

As we discussed in Sec. III, the minimum occurs for positive values of k and is given by

$$k=\frac{H}{3}\left[1+\left[1+\frac{3\gamma}{H^2}\right]^{1/2}\right]\ . \tag{A2}$$

The local minimum of Ψ is

$$\begin{aligned}\Psi_{\min}&=\Psi(k)\\&=-\left[\tfrac{2}{27}H^3+\tfrac{1}{3}\gamma H\right.\\&\qquad\left.+\left(\tfrac{2}{27}H^3+\tfrac{2}{9}\gamma H\right)\left[1+\frac{3\gamma}{H^2}\right]^{1/2}\right]\ .\end{aligned} \tag{A3}$$

We need to show that $\Psi_{\min}\leq-\gamma H$, or equivalently that

$$\tfrac{2}{27}H^3-\tfrac{2}{3}\gamma H+\left(\tfrac{2}{27}H^3+\tfrac{6}{27}\gamma H\right)\left[1+\frac{3\gamma}{H^2}\right]^{1/2}\geq 0\ . \tag{A4}$$

We divide this expression by H, which is positive, and rearrange the resulting expression,

$$9\gamma-H^2\leq H^2\left[1+\frac{3\gamma}{H^2}\right]^{3/2}\ . \tag{A5}$$

To show that this inequality holds, we consider two cases. (i) If $9\gamma\leq H^2$, then $9\gamma-H^2\leq 0<H^2(1+3\gamma/H^2)^{3/2}$ and Eq. (A5) holds. (ii) If $9\gamma\geq H^2$, then we take the square on both sides of Eq. (A5) and after a few simple rearrangements obtain

$$0\leq 27\gamma(\gamma-H^2)^2\ ,$$

which is obviously fulfilled, and implies that Eq. (A5) holds. Thus we find that indeed for all values of the noise parameters $\Psi_{\min}\leq-\gamma H$.

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