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Noise-Induced Clumping in the One-Dimensional Reversible Diffusion-Limited Single-Species Coagulation Process

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We investigate the interplay of internal fluctuations and external noise in the diffusion-reaction system $A + A \leftrightarrow A$, where the coagulation reaction is in the diffusion-controlled regime. Our theoretical treatment of the system is based on a recently developed exact and unified description that accounts for both types of random fluctuations. Specifically, we study the case where the external noise affects the diffusion coefficient, i.e., the coagulation rate, and is given by a dichotomous Markov process. We provide exact solutions for the steady state of the system and show that the spatially homogeneous external noise drives the system out of thermodynamic equilibrium. The noise induces microscopic spatial correlations between the particle positions. We compare this noise-induced clumping to the previously studied case of external noise in the birth rate, and discuss the similarities and differences.

KEY WORDS: Interacting particle system; diffusion-reaction system; external noise; dichotomous Markov process.

1. INTRODUCTION

The effects of external noise on the dynamical behavior of nonlinear systems that lack spatial degrees of freedom are well understood (see, e.g., ref. 1). Random fluctuations in the environment of the system can postpone or advance instabilities, and can even given rise to transitions to states that cannot occur if the surroundings are free of stochastic variations. The influence of external noise on the dynamical behavior of spatially distributed nonlinear systems is less understood and a topic of current interest (see, e.g., ref. 2). External noise can interact with the spatial degrees

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of freedom of the system as well as with the internal fluctuations intrinsic to many-body systems. We expect the latter effect to be particularly important for diffusion-limited reactions in low spatial dimensions. In such systems internal fluctuations can give rise to strong particle–particle correlations that dominate the macroscopic dynamics.[3–6] To describe and analyze the behavior of reaction-diffusion systems with internal fluctuations and external noise is a complicated, and often intractable, task. It is therefore useful and desirable to find a simple many-body model system for which an exact, unified description of internal fluctuations and external noise can be easily formulated and for which exact analytical solutions can be obtained.

One of the simplest diffusion-reaction systems is the reversible coagulation-growth process, $A + A \leftrightarrow A$, with irreversible input, $B \rightarrow A$, in one spatial dimension. We refer to the objects A as "particles" and consider the case where the transport of A constitutes the rate-limiting step for the reaction $A + A \rightarrow A$; the particles coalesce immediately upon contact. In other words, the coagulation reaction is in the *diffusion-controlled regime.* This diffusion-reaction system is a simple interacting-particle system, and its behavior in the absence of external noise has been studied extensively and is well understood.[5,6,8–13] The model has attracted attention because it allows for an exact and closed microscopic formulation in terms of a single linear partial differential equation. In statistical physics many-body systems are typically described with a set of joint probability density functions. This approach encounters the usual closure problem: No finite subset of probability density functions can fully characterize the system, since the temporal evolution of the lower members of the hierarchy depends on the higher particle densities. For the specific model we consider here, an alternate approach based on the probability $E(x, y, t)$ that the interval $[x, y]$, with $x \leqslant y$, is empty at time t, leads to an exact, closed microscopic description. It is usual to define the model initially on a lattice of lattice spacing Δx and then to take the continuum limit. In this limit the empty-interval probability obeys the following linear partial differential equation[14]:

$$\frac{\partial E(x, y, t)}{\partial t} = \frac{\partial}{\partial x} D(x, t) \frac{\partial E}{\partial x} + \frac{\partial}{\partial y} D(y, t) \frac{\partial E}{\partial y} - \frac{1}{2} v(x, t) \frac{\partial E}{\partial x} + \frac{1}{2} v(y, t) \frac{\partial E}{\partial y} - \left[\int_x^y R(z, t)\, dz \right] E(x, y, t) \quad (1.1)$$

with the boundary condition

$$\lim E(x, y, t) = 1 \qquad \text{for} \quad y \downarrow x \quad \text{or} \quad x \uparrow y \quad (1.2)$$

The other boundary conditions, imposed as either $y \to +\infty$ or $x \to -\infty$, depend on the specific conditions at hand. In (1.1), $D(x, t)$ is the diffusion coefficient, $v(x, t)$ is the birth rate, i.e., the rate of the back reaction $A \to A + A$, and $R(x, t)$ is the rate of particle input at point x.

In the following we will consider only the case that the diffusion coefficient D, the birth rate v, and the rate of particle input R are uniform in space and that the system is statistically spatially homogeneous. With $x' = y - x$, Eq. (1.1) reduces then to[6]

$$\partial_t E(x, t) = 2D\, \partial_{xx} E(x, t) + v\, \partial_x E(x, t) - RxE(x, t) \equiv L(D, v, R)\, E(x, t) \tag{1.3}$$

where we dropped the prime on x. The boundary conditions are

$$E(0, t) = 1 \qquad \text{and} \qquad E(\infty, t) = 0 \tag{1.4}$$

Here $E(x, t)$ is the probability that a randomly chosen interval of length x is empty at time t. The probability that a small interval of length dx is occupied equals $1 - E(dx, t)$. Hence the concentration, or density, of particles is defined by

$$c(t) = -\left.\frac{\partial E(x, t)}{\partial x}\right|_{x=0} \tag{1.5}$$

Note that the concentration $c(t)$ is an ensemble average.[14] It does not itself fluctuate, but if fully takes into account all the microscopic fluctuations in the system, i.e., the internal fluctuations, and any correlations which may develop. (We are considering an infinite system and have taken the thermodynamic limit.) Correlations can be characterized by the interparticle distribution function (IPDF) $p(x, t)$, the probability density of finding the nearest particle a distance x from a given particle. The IPDF is related to the empty-interval probability $E(x, t)$ in the following way:

$$c(t)\, p(x, t) = \frac{\partial^2 E(x, t)}{\partial x^2} \tag{1.6}$$

A more detailed derivation of the evolution equations and the above quantities can be found in refs. 6 and 14.

The organization of this paper is as follows: In Section 2 we will briefly summarize the closed, unified description of internal fluctuations and external noise that was developed in ref. 7 for the reversible diffusion-limited coagulation reaction $A + A \leftrightarrow A$ with irreversible input $B \to A$ in opne spatial dimension. The kinetic equation for the strictly reversible reaction-diffusion system, $R=0$, with constant birth rate v and with dichotomous Markov noise in the diffusion coefficient D is formulated in Section 3. The stationary form of the empty-interval probability, of the IPDF, and of the density is derived in Section 4. The Poisson white noise limit is briefly discussed at the end of that section. Section 5 contains concluding remarks and a discussion of some open problems.

2. A UNIFIED DESCRIPTION OF INTERNAL FLUCTUATIONS AND EXTERNAL NOISE

The kinetic equation (1.1), or (1.3), completely describes all internal fluctuations in the diffusion-reaction system ($A+A \leftrightarrow A$, $B \to A$) in the diffusion-controlled regime. In ref. 7 the closed, exact microscopic description of this model system was extended to include also the effects of external noise. Random variations in the surroundings can give rise to fluctuations in the diffusion coefficient D_t, or the birth rate v_t, or the input rate R_t. A unified description that takes into account the internal fluctuations of the reaction-diffusion system as well as the external noise is achieved by a natural extension of the central quantity of our approach, namely the *joint probability* $E(x, D, v, R, t)$, which is defined by

$$E(x, D, v, R, t)\, dD\, dv\, dR = \text{prob}\{\text{interval of length } x \text{ is empty at time } t \\ \textit{and } D_t \in (D, D+dD), \\ \textit{and } v_t \in (v, v+dv), \\ \textit{and } R_t \in (R, R+dR)\} \tag{2.1}$$

This joint probability obeys the kinetic equation

$$\partial_t E(x, D, v, R, t) = [L(D, v, R) + W_D + W_v + W_R]\, E(x, D, v, R, t) \tag{2.2}$$

for statistically independent external fluctuations in the diffusion coefficient, the birth rate, and the rate of particle input. In (2.2) the operators W are the evoluation operators of the various stochastic processes, i.e.,

$$\partial_t p_j = W_j p_j \tag{2.3}$$

where p_j is the probability (or probability density) of the stochastic process j, and $j=D$, v, or R. The boundary conditions for (2.2) are

$$E(0, D, v, R, t) = p_D(D, t)\, p_v(v, t)\, p_R(R, t) \tag{2.4}$$

and

$$E(\infty, D, v, R, t) = 0 \tag{2.5}$$

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

In ref. 7 explicit, exact results are derived for the strictly reversible system, i.e., $R = 0$, with dichotomous Markov noise in the birth rate v, i.e., the rate of the process $A \to A + A$, and constant D. In particular, the case that the birth rate fluctuates between zero and a fixed positive value is studied. It is found that the external noise drives the system out of thermodynamic equilibrium to a stationary nonequilibrium state. In this state, neither the empty interval probability nor the interparticle distribution function is a simple exponential corresponding to a totally random distribution of particles on the line, the maximum-entropy state characteristic of equilibria. The interaction of the internal fluctuations with the spatially homogeneous external noise gives rise to 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. This effect occurs without threshold, and the deviation from the equilibrium state increases smoothly with the strength of the noise. The noise-induced clumping is most pronounced in the Poisson white noise limit of the birth rate fluctuations. Interestingly, noise in the birth rate does not affect the stationary density; it always equals the equilibrium value for a system with the same average birth rate: $c_s = \langle v_t \rangle / 2D$.

In this paper we investigate the effect of spatially homogeneous external noise in the rate of the coagulation process, $A + A \to A$. Again we expect the interaction of internal fluctuations and external noise to induce correlations and clumping in the system. Indeed, when the diffusion coefficient becomes very small, the coagulation process will be dominated by the back process and existing particles should spawn clumps. However, the limit $D \to 0$ is clearly a singular limit for the problem, viz. the highest-derivative term vanishes, contrary to the limit $v \to 0$, and has to be studied carefully. Further, the density is inversely proportional to D and fluctuations in the diffusion coefficient are therefore expected to change the density, contrary to the case of fluctuations in the birth rate.

3. FLUCTUATING COAGULATION RATE: DICHOTOMOUS EXTERNAL NOISE

We model fluctuations in the diffusion coefficient by a dichotomous Markov process, or random telegraph signal. The diffusion coefficient D takes on only two values $D_t \in \{D_-, D_+\}$, and the lifetime of each state is exponentially distributed. The latter is necessary and suffcient for the process to be Markovian. The kinetic equation, or master equation, for the probability of the process D_t reads

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

where $P_- = \text{prob}\{D_t = D_-\}$, $P_+ = \text{prob}\{D_t = D_+\}$, and α and β are the jump frequencies from one state to the other. The stationary probability for dichotomous noise is given by

$$P_{s+} = \frac{\beta}{\gamma}, \qquad P_{s-} = \frac{\alpha}{\gamma} \tag{3.2}$$

with $\gamma = \alpha + \beta$. As usual we will assume that the environment of the system is stationary. Then the external noise is a stationary random process. Its mean value is

$$\langle D_t \rangle = \frac{\alpha D_- + \beta D_+}{\gamma} \equiv D \tag{3.3}$$

and its correlation function is given by

$$\langle D_{t+\tau} D_t \rangle - \langle D_{t+\tau} \rangle \langle D_t \rangle = \frac{\alpha\beta}{\gamma^2} (D_+ - D_-)^2 \exp(-\gamma |\tau|) \tag{3.4}$$

The correlation time of the dichotomous noise is

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

The kinetic equation (2.2) for the reaction-diffusion system with external dichotomous noise in the diffusion coefficient reads

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

where

$$L_\pm = 2D_\pm \partial_{xx} + v\partial_x - Rx \tag{3.7}$$

We rescale space and time,

$$\begin{aligned} x' &= \frac{v}{2D} x, \qquad t' = \frac{v^2}{2D} t, \qquad d_\pm = \frac{D_\pm}{D} \\ R' &= \frac{4D^2}{v^3} R, \qquad \alpha' = \frac{2D}{v^2} \alpha, \qquad \beta' = \frac{2D}{v^2} \beta \end{aligned} \tag{3.8}$$

to obtain the kinetic equations in dimensionless form (dropping the primes)

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

and

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

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

$$\begin{aligned} E_-(0, t) &= P_{s-} = \frac{\alpha}{\gamma}, \qquad E_-(\infty, t) = 0 \\ E_+(0, t) &= P_{s+} = \frac{\beta}{\gamma}, \qquad E_+(\infty, t) = 0 \end{aligned} \tag{3.11}$$

The dimensionless ensemble-averaged concentration is again computed by

$$c(t) = -\left.\frac{\partial E(x, t)}{\partial x}\right|_{x=0} \tag{3.12}$$

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

$$E(x, t) = E_-(x, t) + E_+(x, t) \tag{3.13}$$

The concentration of the equilibrium state in dimensionless variables is $c_{\text{eq}} = 1$.

We simplify the system of coupled kinetic equations (3.9) and (3.10) by using (3.9) to eliminate E_+ in (3.10). After some simple algebra we obtain

$$\begin{aligned} &\partial_{tt} E_- + \gamma \partial_t E_- - (d_+ + d_-)\, \partial_{xxt} E_- - 2\partial_{xt} E_- + 2Rx\, \partial_t E_- \\ &\quad = -d_+ d_- \partial_{xxxx} E_- - (d_+ + d_-)\, \partial_{xxx} E_- + (\gamma - 1)\partial_{xx} E_- + \gamma \partial_x E_- \\ &\qquad + R[(d_+ + d_-)\, x\partial_{xx} E_- + 2(d_+ + x)\, \partial_x E_- + (1 - Rx^2 - \gamma x) E_-] \end{aligned} \tag{3.14}$$

Recall that with the scaling (3.8) the average value of d_t equals one. Thus, if we fix the lower level of the dichotomous noise

$$d_- = \varepsilon \tag{3.15}$$

then the upper level is given by

$$d_+ = \frac{1}{\beta} (\gamma - \alpha\varepsilon) \tag{3.16}$$

4. STEADY-STATE BEHAVIOR: NOISE-INDUCED CLUMPING

In the absence of external noise and in the case of no particle input, $R=0$, the process settles down to an equilibrium state, characterized by an exponentially decreasing empty-interval probability and IPDF[6]:

$$E_{\rm eq}(x) = e^{-c_{\rm eq} x} \tag{4.1}$$

and

$$p_{\rm eq}(x) = c_{\rm eq} e^{-c_{\rm eq} x} \tag{4.2}$$

This IPDF corresponds to a totally random (Poisson) distribution of particles on the line, which is the maximum-entropy state.

The effect of a fluctuating coagulation rate on the equilibrium state can be determined from (3.14) with $\partial_t = 0$ and $R=0$:

$$0 = -d_+ d_- \partial_{xxxx} E_- - (d_+ + d_-)\partial_{xxx} E_- + (\gamma - 1)\,\partial_{xx} E_- + \gamma \partial_x E_- \tag{4.3}$$

This is a linear differential equation with constant coefficients and it can be solved with the usual ansatz:

$$E_- \propto e^{-kx} \tag{4.4}$$

We obtain the characteristic polynomial

$$0 = -d_+ d_- k^4 + (d_+ + d_-)k^3 + (\gamma - 1)k^2 - \gamma k \tag{4.5}$$

In addition to the eigenvalue $k_1 = 0$, which corresponds to an empty system, we have the roots of the cubic polynomial

$$d_+ d_- k^3 - (d_+ + d_-)k^2 - (\gamma - 1)k + \gamma = 0 \tag{4.6}$$

Equation (4.6) has one negative and two positive real roots. Only the latter are acceptable in light of the boundary condition (3.11) as $x \to \infty$. In the general case the complicated explicit form of the roots is not very enlightening and will not be given here. In the case $d_- = \varepsilon \ll 1$, the case we expect to display the strongest interaction of external noise with internal fluctuations, we can obtain compact analytical expressions for the roots. Making the ansatz $k = \kappa_1/\varepsilon + \kappa_2 + \ldots$ and using (3.16), we find to dominant order:

$$k_2 = \frac{1}{\varepsilon} + \beta + O(\varepsilon) \tag{4.7}$$

and

$$k_{3,4} = \frac{\beta(\gamma - 1)}{2\gamma}\left[-1 \pm \left(1 + \frac{4\gamma^2}{\beta(\gamma-1)^2}\right)^{1/2}\right] + O(\varepsilon) \tag{4.8}$$

Note that k_4 is negative and thus not compatible with the boundary conditions.

Let k_2 and k_3 be the two positive roots of (4.6). (If $d_- = \varepsilon \ll 1$, they are given by the above expressions. Otherwise, the values for k_2 and k_3 are obtained numerically.) The steady-state expression for E_- is given by

$$E_-(x) = C_2 e^{-k_2 x} + C_3 e^{-k_3 x} \tag{4.9}$$

and from (3.9) we obtain the corresponding expression for E_+:

$$\alpha E_+(x) = (-\varepsilon k_2^2 + k_2 + \beta)\, C_2 e^{-k_2 x} + (-\varepsilon k_3^2 + k_3 + \beta)\, C_3 e^{-k_3 x} \tag{4.10}$$

(Here and in the following $\varepsilon < 1$, but not necessarily $\varepsilon \ll 1$.) The coefficients C_2 and C_3 are determined by the boundary conditions at $x = 0$ [see (3.11)], and after some algebra we find

$$C_2 = \frac{\alpha}{\gamma} \frac{k_3(1-\varepsilon k_3)}{k_3(1-\varepsilon k_3) - k_2(1-\varepsilon k_2)} \tag{4.11}$$

and

$$C_3 = -\frac{\alpha}{\gamma} \frac{k_2(1-\varepsilon k_2)}{k_3(1-\varepsilon k_3) - k_2(1-\varepsilon k_2)} \tag{4.12}$$

The stationary empty-interval probability $E(x) = E_-(x) + E_+(x)$ is then given by

$$E(x) = N\{[\gamma + k_2(1-\varepsilon k_2)]\, k_3(1-\varepsilon k_3) e^{-k_2 x} - [\gamma + k_3(1-\varepsilon k_3)]\, k_2(1-\varepsilon k_2) e^{-k_3 x}\} \tag{4.13}$$

with

$$N = \frac{1}{\gamma[k_3(1-\varepsilon k_3) - k_2(1-\varepsilon k_2)]} \tag{4.14}$$

Also we have

$$\frac{\partial E(x)}{\partial x} = N\{-[\gamma + k_2(1-\varepsilon k_2)]\, k_2 k_3(1-\varepsilon k_3) e^{-k_2 x} + [\gamma + k_3(1-\varepsilon k_3)] k_2 k_3(1-\varepsilon k_2) e^{-k_3 x}\} \tag{4.15}$$

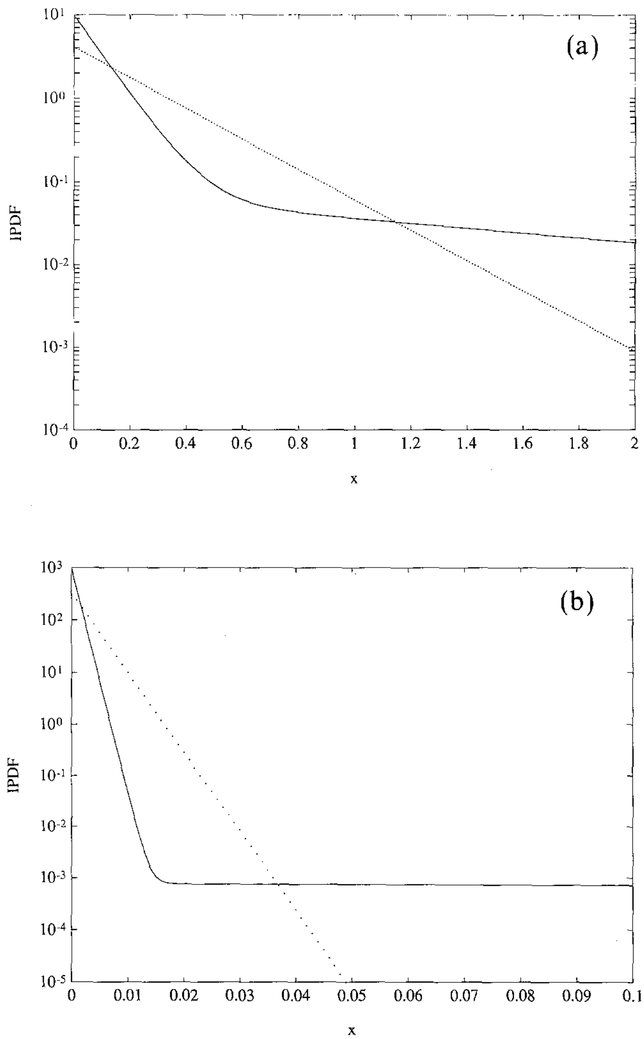
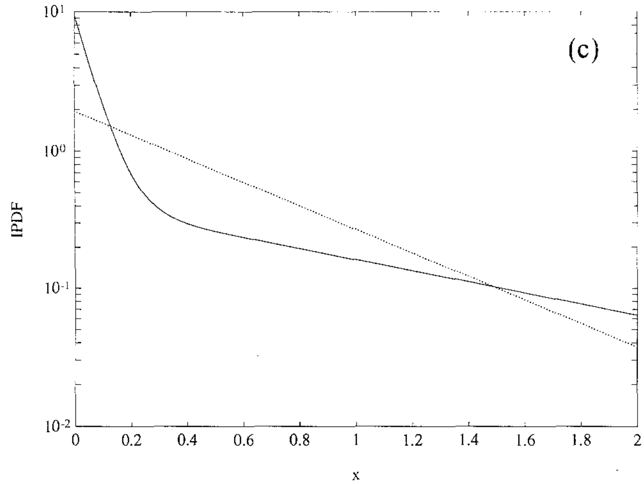


Fig. 1. Semilogarithmic plot of the interparticle distribution function (IPDF) for the noisy system (continuous curve) and an equilibrium system with the same concentration (dotted curve). The noise parameters are: (a) $d_- = 0.1$, $\beta = 1$, $\gamma = 10$; (b) $d_- = 0.001$, $\beta = 1$, $\gamma = 10$; (c) $d_- = 0.1$, $\beta = 10$, $\gamma = 100$; (d) $d_- = 0.001$, $\beta = 10$, $\gamma = 100$. Note the change in the scale of the x axis between (a) and (b) and between (c) and (d). The stationary densities are: (a) $c_s = 4.22$; (b) $c_s = 354$; (c) $c_s = 1.98$; (d) $c_s = 77.7$.



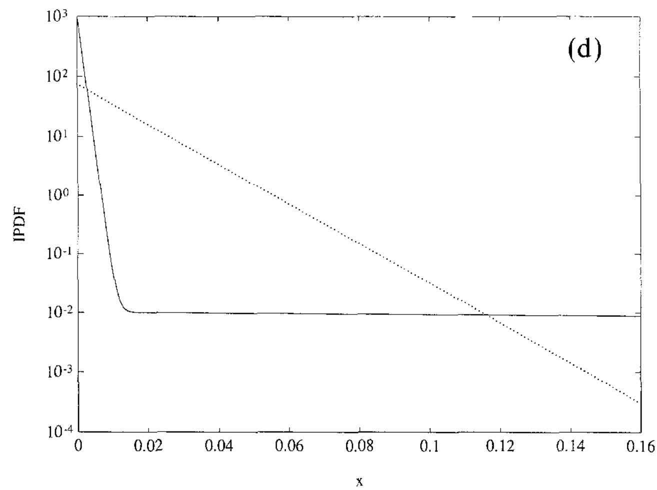


Fig. 1. (*Continued*)

the stationary density

$$c_s = -\frac{\partial E(x, t)}{\partial x}\bigg|_{x=0} = N(k_3 - k_2)[\gamma k_2 k_3 \varepsilon + k_2 k_3 (1 - \varepsilon k_2)(1 - \varepsilon k_3)] \tag{4.16}$$

(which does *not* equal $\langle 1/d_t \rangle$, or $\langle v/2D_t \rangle$ in dimensioned variables), and the stationary IPDF

$$p_s(x) = \frac{1}{c_s} \frac{\partial^2 E(x)}{\partial x^2}$$
$$= \frac{1}{c_s} N\{[\gamma + k_2(1 - \varepsilon k_2)] k_2^2 k_3(1 - \varepsilon k_3) e^{-k_2 x}$$
$$- [\gamma + k_3(1 - \varepsilon k_3)] k_2 k_2^3 (1 - \varepsilon k_2) e^{-k_3 x}\} \tag{4.17}$$

For $\varepsilon \ll 1$ the expression for the stationary density reduces to

$$c_{s,\text{asym}} = \frac{1}{\varepsilon\gamma} \frac{k_3 \alpha}{k_3 + \beta} + O(\varepsilon^0) \tag{4.18}$$

and that of the IPDF to

$$p_{s,\text{asym}}(x) = \frac{1}{\varepsilon} \exp\left(-\frac{x}{\varepsilon}\right) + O(\varepsilon^0) \tag{4.19}$$

where k_3 is given by (4.8).

As expected, macroscopic external fluctuations in the diffusion coefficient, or in other words in the coagulation rate, drive the system out of equilibrium. They drive the IPDF, as well as the empty-interval probability, away from the simple exponential distribution. The form of the IPDF (4.17) indicates that the interaction of the *spatially homogeneous* external noise with the statistically homogeneous internal fluctuations induces microscopic particle–particle correlations, clumping, and destroys the property of detailed balance. The external noise induces a *nonequilibrium* steady state in the strictly reversible diffusion-reaction system. The noise-induced clumping is clearly revealed in Fig. 1, where we plot $p_s(x)$ vs. x for several values of d_-, β, and γ. The equilibrium IPDF for a system with the same density as the noisy system is plotted for comparison. The nonequilibrium $p_s(x)$ is larger than the equilibrium $p_{\text{eq}}(x)$ for small x and for large x. This indicates that the particles bunch together; the system displays relatively more of the smaller and of the larger gaps between adjacent particles as compared to the equilibrium state of a completely independent distribution of particles.

The first term in the IPDF approaches ever more closely a Dirac δ-distribution as $d_- = \varepsilon$ approaches zero. This behavior is independent of the correlation time of the noise and occurs already for finite γ. This is in contrast to the case of dichotomous Markov noise in the birth rate. The IPDF displays a δ-peak at zero only as v_+ approaches infinity, which in turn requires that γ diverges.[7] In other words, the birth-rate fluctuations must approach the white noise limit. The limit of vanishing correlation time, $\tau_{\text{cor}} \to 0$, does not give rise to new behavior for external noise in the diffusion coefficient. The Poisson white noise limit of the dichotomous Markov process corresponds to[15]

$$d_+ \to \infty, \quad \alpha \to \infty, \qquad \text{such that} \quad \frac{d_+}{\alpha} = \sigma = O(1) \tag{4.20}$$

Using (3.16), we obtain

$$\sigma\beta = 1 - \varepsilon \tag{4.21}$$

As the Poisson white noise limit is approached, we keep only the dominant terms in the coefficients of the characteristic polynomial (4.6), which reduces to

$$\alpha\sigma(1-\beta\sigma)k^3 - \alpha\sigma k^2 - \alpha k + \alpha = 0 \tag{4.22}$$

or

$$\sigma(1-\beta\sigma)k^3 - \sigma k^2 - k + 1 = 0 \tag{4.23}$$

The structure of (4.22) differs from that of the corresponding characteristic polynomial for the fluctuating birth-rate case. There the coefficient of the cubic term contains no α, while the coefficients of all other terms are proportional to α. This results in an eigenvalue $k \propto \alpha$, and hence a contribution to the IPDF that approaches a Dirac δ-function in the white noise limit. In our case, there is no eigenvalue that approaches infinity as α approaches infinity. As in the case of finite correlation times, (4.22), or (4.23), has a root that diverges if the lower level of the noise approaches zero, i.e.,

$$\varepsilon = 1 - \beta\sigma \to 0$$

5. DISCUSSION

We have shown that spatially homogeneous, macroscopic external noise in the diffusion coefficient of the reversible diffusion-limited coagulation reaction $A + A \leftrightarrow A$ drives the system out of thermodynamic equilibrium. The noise interacts with the internal fluctuations of the system and the spatial degrees of freedom to induce a nonequilibrium steady state in which the particle positions are correlated. The particles tend to bunch together on average. This effect occurs already for finite correlation times of the noise and becomes more pronounced as the lower level of the dichotomous noise decreases, $d_- \to 0$, and as the lifetime of the level increases, $\beta \to 0$. This clumping phenomenon shares some features with the clumping induced by birth-rate fluctuations in the same system. The main differences are that fluctuations in diffusivity change the stationary density —it increases—and produce a quasi-Dirac-δ-function peak in the interparticle distribution function already for colored noise, if the diffusion coefficient becomes small during random time intervals.

There remain several interesting questions: How do fluctuations in the birth rate or the diffusion coefficient affect the dynamic transition in the relaxation kinetics of the reversible process $A + A \leftrightarrow A$? This transition occurs in the absence of random variations in the surroundings when switching between equilibria of different values of the system parameters. How does the clumping depend on the character of the noise, i.e., its state space and probability distribution? How does spatially inhomogeneous noise, for instance, spatial disorder frozen in time, affect the equilibrium state of the diffusion-reaction system? Work on these questions is currently in progress and results will be reported elsewhere.

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