# The Effect of External Noise on Dynamic Behaviors of the Schlögl Model with the First Order Transition for a Photochemical Reaction

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The Schlögl model with the first order transition for a photochemical reaction is considered to study the dynamic behaviors in the neighborhood of the Gaussian white noise by obtaining the explicit results of the time-dependent variance and time correlation function with the aid of approximate methods based on the stationary properties of the system. Then, we discuss the effect of external noise strength on the stability of the model at steady states in detail.

#### Introduction

Since Kuznetsov et al. studied the nonnegligible effect of the external noise in the valve oscillator,1 theoretical and experimental studies of external noise situations have been reported in the various systems, such as nematic liquid crystals,2 dye laser system,3 nonlinear electric circuits,4~6 chemical reactions<sup>7-8</sup> and hydrodynamic instabilities.<sup>9</sup> External noise refers fluctuations present in a given system which are not self-originating. External fluctuations exist when the system is placed in a stochastic environment or when it is stochastically driven by the controlled fluctuations of one of parameters. The mathematical modeling of external noise is made of by considering a deterministic equation appropriate in the absence of external fluctuations. One then considers the external parameter which undergoes fluctuations to be a stochastic variable. The noise term of the stochastic differential equation obtained in this way is usually of multiplicative character, that is, it depends on the instantaneous value of the variables of this system. It does not scale with the size of system and is not necessarily small. We may consider the external noise as an external field which drives the system.

An external noise is frequently assumed to satisfy the Ornstein-Uhlenbeck process, which is only stationary Markov stochastic process. <sup>10-12</sup> The Gaussian white noise is obtained from the Ornstein-Uhlenbeck process by reducing the controllable correlation time of the noise, which is the parameter independent of the noise intensity, to zero. <sup>10-11</sup> For the sake of mathematical convenience the Gaussian white noise is most frequently applied in the theoretical stochastic process.

An external noise plays an important role in the nonequilibrium system: this can postpone or advance instabilities, and may even give rise to the shift of bifurcation diagram and transitions to states that cannot occur if the surroundings are free from random fluctuations. Such phenomena are interpreted theoretically as being associated with changes that the stationary probability distribution of the system undergoes when the noise parameters are varied, <sup>13-20</sup> even though the general validity of this indirect interpretation is questioned.

We consider the Schlögl model with the first order transi-

tion for a photochemical reaction. It is assumed that the system is spatially homogeneous and the size of the system is large enough to neglect internal fluctuations. The purpose of the present paper is to investigate the effect of the external fluctuating light intensity on the stability of the steady states in the neighborhood of the Gaussian white noise by obtaining the explicit results of the time-dependent variance and time correlation function with the aid of approximate methods based on the stationary properties of the system. The approximate method at the unstable steady state is different from that at the stable steady state. The result at the unstable steady state will directly shows that the strength of the external noise stabilizes the unstable steady state.

In the next section the Fokker-Planck equation near the region of the Gaussian white noise is obtained with the aid of the wide band perturbation method. 10~11 By using the equation we discuss the effect of external light on the stationary properties of the system at the steady states. With approximate methods based on the stationary properties we obtain time-dependent variance and correlation function at the stable and unstable steady states. In the final section we discuss the results of the present work.

#### Theory

Let us consider the Schlögl model with the first order transition for a photochemical reaction<sup>21</sup>

$$A + X \xrightarrow{k_1} 3X + B, \ X \xrightarrow{h\nu} C,$$
 (1)

where  $k_1$  and  $k_{-1}$  are the rate constants, A is the reactant and B and C denote the products. The rate equation for the concentration of the intermediate X is given by the following equation with the concentrations of A and B being held constants

$$\frac{\mathrm{d}X}{\mathrm{d}t} = -X^3 + \gamma X - I(1 - \exp(-\alpha X); \ \gamma = \frac{k_1 A}{k_{-1} B},$$

$$I = \frac{I_o}{k_{-1} B}, \ t = k_{-1} B \tau. \tag{2}$$

In Eq. (2), X denotes the concentration of the intermediate;

 $\tau$  is the real time;  $I_0$  is the incident light intensity and  $\alpha$  is the absorption coefficient times the sample thickness. It is assumed that the intensity satisfies the Ornstein-Uhlenbeck process

$$\frac{d}{dt}I(t) = -\lambda I(t) + \sigma \xi(t), \tag{3}$$

where  $\lambda > 0$ ,  $\xi(t)$  is the external noise and  $\sigma$  is the strength of the noise. The noise satisfies the Gaussian condition

$$\langle \xi(t) \rangle = 0, \ \langle \xi(t)\xi(t') \rangle = \delta(t-t').$$
 (4)

In Eq. (4)  $\delta(t-t')$  is the Dirac delta function. The steady state values of X for small extinction coefficient are

$$X_s = 0, \pm (\gamma - \alpha I_s)^{1/2}.$$
 (5)

When  $I_s > \gamma/\alpha$ , there exists only one stable steady state,  $X_s = 0$ . In the case of  $I_s < \gamma/\alpha$ , there are three steady states, that is,  $X_s = 0$  and  $\pm (\gamma - \alpha I_s)^{1/2}$  corresponding to the unstable and stable steady states, respectively.

Defining the fluctuating parts of X and I from the steady state values due to the noise

$$x(t) = X(t) - X_s, i(t) = I(t) - I_s,$$
 (6)

the equations for x(t) and i(t) become

$$\frac{d}{dt}x(t) = (\gamma - 3X_s^2 - \alpha I_s)x(t) - 3X_sx(t)^2 - x(t)^3 - \alpha [X_s + x(t)]i(t),$$

$$\frac{d}{dt}i(t) = -\lambda i(t) + \sigma \xi(t). \tag{7}$$

According to the Wiener-Khintchine theorem<sup>12</sup> the spectral density of the Ornstein-Uhlenbeck process is

$$S^{i}(\omega) = \langle i(\omega)i(\omega)^{\bullet} \rangle = \frac{2\pi\sigma^{2}}{\omega^{2} + \lambda^{2}}, \tag{8}$$

where

$$i(\omega) = \int_{-\infty}^{\infty} \exp(-i\omega t) i(t) dt, \qquad (9)$$

Considering the linear parts of Eq. (7), the time correlation functions are

$$G_o(t) = \langle x(t)x(0) \rangle = \langle x(0)^2 \rangle \exp -|t|/t_{mac},$$

$$G_o'(t) = \langle i(t)i(0) \rangle = \frac{\sigma^2}{\lambda} \exp -|t|/t_{noise},$$
(10)

where we have assumed that  $\langle x(t)i(t')\rangle = 0$  and  $\langle i(t)\xi(t')\rangle = 0$ . The correlation times for the fluctuating macroscopic variable and intensity are given by

$$t_{mac} = \frac{1}{3X_S^2 - \gamma + \alpha I_S}, \ t_{noise} = \frac{1}{\lambda} = \varepsilon^2, \ (\varepsilon^2 < 1).$$
 (11)

Substituting  $\lambda = \epsilon^{-2}$  into Eq. (8) and transforming into  $\sigma/\epsilon$ , the spectral density may be written as

$$S^{i}(\omega) = \langle i(\omega)i(\omega)^{*} \rangle = \frac{2\pi\sigma^{2}}{\varepsilon^{2}\omega^{2} + \varepsilon^{\frac{1}{2}}}$$
 (12)

In the limit  $\varepsilon \to 0$  the correlation time  $t_{noise}$  tends to zero and the bandwidth goes to infinity. Therefore, the spectrum becomes flat, but the spectral density vanishes for all finite frequencies. This is noiseless limit. To avoid ending up with

a noiseless limit and to obtain the correct white noise limit, the intensity of external fluctuations should be appropriately scaled in Eq. (7). Scaling i by  $i/\varepsilon$ , the spectral density is

$$S^{i/\varepsilon}(\omega) = \langle \frac{i(\omega)}{\varepsilon} \frac{i(\omega)^*}{\varepsilon} \rangle = \frac{2\pi\sigma^2}{\varepsilon^4\omega^2 + 1}$$
 (13)

The above spectral density has the finite value of  $2\pi\sigma^2$  in the limit  $\varepsilon \to 0$  called as the Gaussian white noise limit.

Since our main interest is to investigate dynamic behaviors near the region of the Gaussian white noise, Eq. (7) can be rewritten in the neighborhood of the white noise

$$\frac{d}{dt}x(t) = f(x) + \frac{g(x)}{\varepsilon} \quad i(t), \quad \frac{d}{dt}i(t) = -\frac{1}{\varepsilon^2}i(t) + \frac{\sigma}{\varepsilon}\xi(t), \quad (14)$$

where

$$f(x) = (\gamma - 3X_s^2 - \alpha I_s)x - 3X_s x^2 - x^3, \ g(x) = -\alpha (X_s + x). \tag{15}$$

The Fokker-Planck equation corresponding to the above Langevin equation is

$$\frac{\partial}{\partial t} P^{\varepsilon}(x,i,t) = -\frac{\partial}{\partial x} \left[ f(x) + \frac{1}{\varepsilon} g(x) i(t) \right] P^{\varepsilon}(x,i,t) 
+ \frac{1}{\varepsilon^{2}} \frac{\partial}{\partial i} i(t) P^{\varepsilon}(x,i,t) + \frac{\sigma^{2}}{2\varepsilon^{2}} \frac{\partial^{2}}{\partial i^{2}} P^{\varepsilon}(x,i,t).$$
(16)

The exact solution for the nonlinear Fokker-Planck equation is not available. Thus, using the wide band perturbation expansion  $^{10\sim11}$  and taking the result up to the  $\epsilon^2$  term, we have

$$P^{\epsilon}(x,i,t) = P(x,t)P(i); \tag{17}$$

$$\frac{\partial}{\partial t} P(i) = -\frac{\partial}{\partial i} \left[ i(t)P(i) \right] + \frac{\sigma^2}{2} \frac{\partial^2}{\partial i^2} P(i) = 0, \quad (18a)$$

$$\frac{\partial}{\partial t} P(x,t) = \left[ -\frac{\partial}{\partial x} f(x) + \frac{\sigma^2}{2} \frac{\partial}{\partial x} g(x) \frac{\partial}{\partial x} h(x) \right] P(x,t);$$

$$h(x) = g(x) - \varepsilon^2 [g(x)'f(x) - f(x)'g(x)]. \tag{18b}$$

The above equations show that the noise is in the stationary state and the macroscopic variable in the Gaussian white noise limit satisfies the Stratonovich stochastic process, that is, in the  $\varepsilon \to 0$  limit the Fokker-Planck equation corresponds to the Stratonovich definition of the following Langevin equation with multiplicative noise<sup>12</sup>

$$\frac{d}{dt}x(t) = f(x) + \sigma g(x)\xi(t). \tag{19}$$

At first, let us consider the static properties of the model in order to discuss the nonlinear effect on the time correlation function. Considering the  $\varepsilon^2$  term as the perturbed term, the stationary solution of Eq. (18) may be written as

$$P_{s}(x) = P_{os}(x) \left\{ 1 + \varepsilon^{2} \left[ C + \frac{g(x)'f(x) - f(x)'g(x)}{g(x)} - \frac{f(x)^{2}}{\sigma^{2} g(x)^{2}} \right] \right\}, \quad (20)$$

where

$$C = \int_{-\infty}^{\infty} P_{os}(x) \left\{ \frac{f(x)'g(x) - f(x)g(x)'}{g(x)} - \frac{f(x)^2}{\sigma^2 g(x)^2} \right\} dx \qquad (21)$$

$$P_{os}(x) = Ng(x)^{-1} \exp\left[\frac{2}{\sigma^2}\int_{-\pi}^{x} \frac{f(y)}{g(y)^2} dy\right]. \tag{22}$$

In Eq. (22) N is the normalization constant.

At the stable steady state  $P_{ns}(x)$  is assumed to be Gaussian, that is,

$$P_{os}(x) = \left(\frac{2}{\pi \alpha^2 \sigma^2}\right)^{1/2} \exp{-\frac{2x^2}{\alpha^2 \sigma^2}}.$$
 (23)

 $P_s(x)$  is up to the second order of x

$$P_{s}(x) = P_{os}(x) \left( 1 + \varepsilon^{2} \left\{ \frac{27}{8} \alpha^{2} \sigma^{2} + 8X_{s} x - \frac{2}{\alpha^{2} \sigma^{2}} \left[ \alpha^{2} \sigma^{2} + 2(\gamma - \alpha I_{s}) x^{2} \right] \right\} \right). \tag{24}$$

For the case of  $\varepsilon < 1$  we may neglect the perturbed term in Eq. (24). The variance and nth order moments are

$$\langle x^2 \rangle_s = \int_{-\infty}^{\infty} x^2 P_{vs}(x) dx = \frac{\alpha^2 \sigma^2}{4},$$
 (25a)

 $\langle x^n \rangle_s = \begin{bmatrix} 0, & \text{if } n \text{ is an odd positive integer,} \\ \text{all possible pair products, if n is an even positive integer.} \end{bmatrix}$  (25b)

There is great interest in the unstable steady state,  $X_s = 0$ . The stationary probability distribution is

$$P_{ns}(x) = \frac{1}{(\alpha\sigma)^{\beta}\Gamma(\beta/2)} |x|^{\beta-1} \exp{-\frac{x^2}{\alpha^2\sigma^2}}; \ \beta = \frac{2(\gamma - \alpha I_s)}{\alpha^2\sigma^2} > 0,$$
(26)

where  $\Gamma(\beta/2)$  is the gamma function. In the case of the Gaussian white noise limit ( $\varepsilon=0$ ), the dependence of the stationary probability distribution on the light intensity and noise strength is shown in Figure 1a. The figure shows that the parameters affect the state of the system profoundly. When  $\beta>1$ , the probability distribution is a binodal Gaussian distribution with the maximum at  $x=\pm \left[\gamma-\alpha I_s-\alpha^2\sigma^2/2\right]^{1/2}$ . The maximal peaks show that the deterministic stable steady states are shifted to  $\pm \left[\gamma-\alpha I_s-\alpha^2\sigma^2/2\right]^{1/2}$  due to the external noise. When  $\beta=1$  and <1, the distribution is a Gaussian and a delta-like distribution, respectively. Including the perturbed term,  $P_s(x)$  is  $^{22}$ 

$$P_{s}(x) = P_{os}(x) \left\{ 1 - \varepsilon^{2} \left[ 3(\gamma - \alpha I_{s}) - 4x^{2} + \frac{1}{\alpha^{2} \sigma^{2}} (\gamma - \alpha I_{s} - x^{2})^{2} \right] \right\} (27)$$

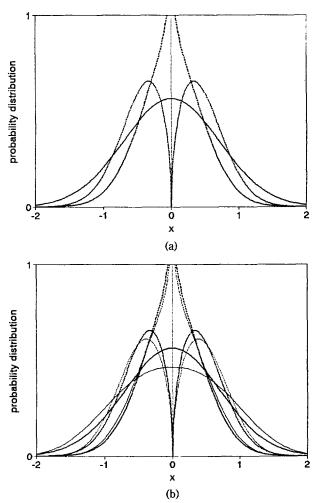
In Eq. (27) the perturbed term should be less than unity. As  $\epsilon$  increases, the probability distribution deviates from the case of  $\epsilon = 0$ . As shown in Figure 1b, the distribution of  $\epsilon = 0.10$  corresponds to that of  $\epsilon = 0$ . When  $\epsilon = 0.31$ , it slightly deviates from that of  $\epsilon = 0$ . Thus, when  $\epsilon < 1$ , the term including  $\epsilon^2$  may be neglected. The variance and nth order moments are

$$\langle x^2 \rangle_s = \int_{-\infty}^{\infty} x^2 P_{os}(x) dx = \gamma - \alpha I_s,$$
 (28)

$$\langle x^{2n+1}\rangle_s = 0, \ \langle x^{2n}\rangle_{k=0} = \prod^{n-1} \langle x^2 + k\alpha^2\sigma^2\rangle,$$
 (29)

if n is a positive integer.

As shown in Eq. (10), the time correlation function between the fluctuating macroscopic variables does not include the noise effect at the steady states and especially diverges at the unstable steady state. As previously stated, the state of the system at  $X_s=0$  is severely influenced by the noise strength. Thus, let us obtain the correlation function from the nonlinear equation.



**Figure. 1.** (a) The stationary probability distribution of Eq. (26). The heavy dotted, heavy solid and heavy dashed lines denote the distribution of  $\beta=1.47$ , 1 and 0.84, respectively. The values of parameters, when  $\beta=1.47$ , are  $\gamma=2$ ,  $\alpha=0.3$ ,  $I_s=5.5$  and  $\sigma=2.3$ . In the case of  $\beta=1$ ,  $\gamma=2$ ,  $\alpha=0.5$ ,  $I_s=3$  and  $\sigma=2.$  When  $\beta=0.84$ ,  $\gamma=2$ ,  $\alpha=0.3$ ,  $I_s=6.0$  and  $\sigma=2.3$ . (b) The dependence of the probability distributions on the parameter,  $\varepsilon$ . The heavy and light lines denote  $\varepsilon=0$  and 0.31, respectively. The lines of =0.1 correspond to those of  $\varepsilon=0$ . The values of other parameters are the same as in Figure 1a.

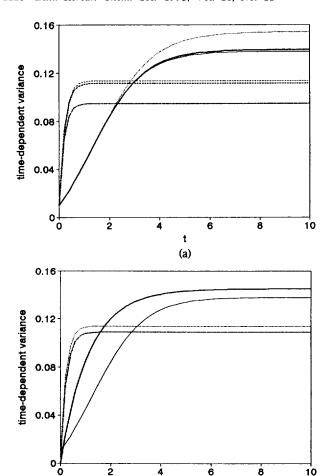
Using Eq. (7), the time correlation function satisfies the following equation

$$\frac{d}{dt}G(t) = (\gamma - 3X_s^2 - \alpha I_s)G(t) - \langle x(t)^3 x(0) \rangle, \tag{30}$$

With the aid of Eq. (18b) the time-dependent variance is

$$\frac{d}{dt} \langle x(t)^2 \rangle = \sigma^2 \alpha^2 X_s^2 + 2\{ (\gamma - 3X_s^2 - \alpha I_s) + \alpha^2 \sigma^2 [1 + \varepsilon^2 (\gamma - 12X_s^2 - \alpha I_s)] \} \langle x(t)^2 \rangle - 2(1 - 4\varepsilon^2 \alpha^2 \sigma^2) \langle x(t)^4 \rangle.$$
(31)

Since it is not available to obtain the exact solutions for the time correlation function and variance, an approximate method is introduced. The approximate method at the unstable steady state is different from that at the stable steady state. Thus, dynamic behaviors at the stable and unstable



**Figure. 2.** (a) The time-dependent variance at the stable steady state. The dashed, heavy dashed and heavy dotted lines represent  $\varepsilon=0$ , 0.10 and 0.31 for 2  $(\gamma-\alpha I_s)>\alpha^2\sigma^2$ , respectively. The solid, heavy solid and dotted lines denote  $\varepsilon=0$ , 0.10 and 0.31 for  $2(\gamma-\alpha I_s)<\alpha^2\sigma^2$ , respectively. The values of other parameters are  $\gamma=2$ ,  $\alpha=0.3$ ,  $\langle x(0)^2\rangle=0.01$  and  $\alpha=2.2$ . The light intensity  $I_s=3$  (and 6) is taken for  $2(\gamma-\alpha I_s)>\alpha^2\sigma^2$  (and  $2(\gamma-\alpha I_s)<\alpha^2\sigma^2$ ), respectively. The same values of the parameters are used in Figs. 2b and 3. (b) The comparison of the variances at the stable steady state given in Eqs. (34) and (36), when  $\varepsilon=0$ . The results of Eq. (34) for  $2(\gamma-\alpha I_s)>$  (and  $<)\alpha^2\sigma^2$  are represented by the dotted and solid lines. The other heavy lines denotes the approximate results of Eqs. (36).

t

(b)

steady states are discussed separately.

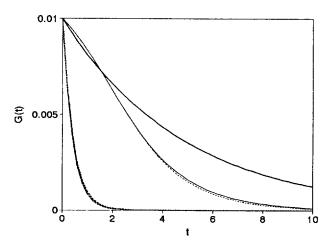
Let us first consider the Gaussian approximation at the stable steady state and Eqs. (30) and (31) are reduced to

$$\frac{d}{dt}G(t) = -\left[2(\gamma - \alpha I_s) + 3\langle x(t)^2 \rangle\right]G(t), \tag{32}$$

$$\frac{d}{dt} \langle x(t)^2 \rangle = \alpha^2 \sigma^2 (\gamma - \alpha I_s) + 2\{-2(\gamma - \alpha I_s) + \alpha^2 \sigma^2 [1 - 11\epsilon^2 \times (\gamma - \alpha I_s)]\} \langle x(t)^2 \rangle - 6(1 - 4\epsilon^2 \alpha^2 \sigma^2) \langle x(t)^2 \rangle^2.$$
 (33)

The explicit result of the variance is

$$\langle x(t)^2 \rangle = \frac{D[E + 2a\langle x(0)^2 \rangle] \exp\sqrt{\Delta t} - E[D + 2a\langle x(0)^2 \rangle]}{2a[D + 2a\langle x(0)^2 \rangle - (E + 2a\langle x(0)^2 \rangle) \exp\sqrt{\Delta t}]}, \quad (34)$$



**Figure. 3.** The comparison of the time correlation functions at the stable steady state given in Eqs. (37) and (38). When  $2(\gamma - \alpha I_s) > \alpha^2 \sigma^2$ , the result of Eq. (38a) corresponds to with Eq. (37) for the cases of  $\epsilon = 0$ , and  $\epsilon = 0.31$ . In the case of  $2(\gamma - \alpha I_s) < \alpha \sigma^2$ , the result of Eq. (38b) agrees well with Eq. (37) for the case of  $\epsilon = 0$ . For  $\epsilon = 0.31$  in the case of  $2(\gamma - \alpha I_s) < \alpha^2 \sigma^2$  the function deviates from that of  $\epsilon = 0$ .

$$a = -6(1 - 4\varepsilon^2 \alpha^2 \sigma^2), \quad b = -4(\gamma - \alpha I_s) + 2\alpha^2 \sigma^2 [1 - 11\varepsilon^2 (\gamma - \alpha I_s)],$$

$$c = \alpha^2 \sigma^2 (\gamma - \alpha I_s), \tag{35}$$

$$D=b+\sqrt{\Delta}$$
,  $E=b-\sqrt{\Delta}$ ,  $\Delta=b^2-4ac$ .

The example of the dependence of  $\langle x(t)^2 \rangle$  on the parameters are shown in Figure 2a. The variance in the case of  $\gamma$ - $\alpha I_s > \alpha^2$  approaches to its stationary value more rapidly than that of  $\gamma$ - $\alpha I_s < \alpha^2 \sigma^2$ . When  $\varepsilon = 0.10$ , the variances are almost the same as those of  $\varepsilon = 0$ . As  $\varepsilon$  increases, the variance deviates from the case of  $\varepsilon = 0$ . The results in the two cases are

$$\langle x(t)^2 \rangle \simeq \begin{bmatrix} \frac{\alpha^2 \sigma^2}{4} [1 - \exp(-4(\gamma - \alpha I_s)t)], & \text{if } 2(\gamma - \alpha I_s) > \alpha^2 \sigma^2, \quad (36a) \\ \frac{\alpha^2 \sigma^2}{3} [1 - \exp(-2\alpha^2 \sigma^2 t)], & \text{if } 2(\gamma - \alpha I_s) < \alpha^2 \sigma^2. \quad (36b) \end{bmatrix}$$

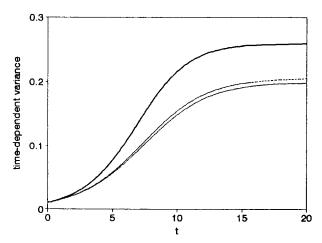
The result of Eq. (36a) indicates that the deterministic and noise terms play the major and minor roles, respectively. However, Eq. (36b) shows that the very strong noise destroys the order based on the deterministic equation. As  $t\rightarrow\infty$ , Eq. (36a) corresponds to Eq. (25a), since the stochastic process is based on the deterministic rate. Accordingly, we may say that the approximation method is correct after long time. The comparison between the results of Eqs. (33) and (35) is given in Figure 2b. Substituting Eq. (34) into Eq. (36), we have

G(t) =

$$\langle x(0)^{2} \rangle \left[ \frac{D + 2a \langle x(0)^{2} \rangle - (E + 2a \langle x(0)^{2} \rangle) \exp\sqrt{\Delta} \ t}{D - E} \right]^{3(E - D)/2a\sqrt{\Delta}}$$

$$\times \exp{-2\left(\gamma - \alpha I_{s} + \frac{3E}{4a}\right)} t. \tag{37}$$

The results in the two cases are



**Figure. 4.** The dependence of variance at the unstable steady state on  $\varepsilon$ . The solid, dashed and heavy solid lines represents  $\varepsilon = 0$ , 0.10 and 0.31, respectively. The values of other parameters are  $\gamma = 2$ ,  $\alpha = 0.3$ ,  $I_s = 6$ ,  $\langle x(0)^2 \rangle = 0.01$  and  $\sigma = 1.7$ .

$$G(t) \simeq \left[ \frac{\langle x(0)^2 \rangle \exp{-2(\gamma - \alpha I_s + \frac{1}{8}\alpha^2 \sigma^2)t}, \text{ if } 2(\gamma - \alpha I_s) > \alpha^2 \sigma^2, (38a)}{\langle x(0)^2 \rangle \left[ \frac{2\alpha^2 \sigma^2}{3(\gamma - \alpha I_s)} \right]^{1/2} \exp{-2(\gamma - \alpha I_s + \frac{1}{2}\alpha^2 \sigma^2)t},$$
if  $2(\gamma - \alpha I_s) < \alpha^2 \sigma^2.$  (38b)

The comparison between the results given in Eqs. (37) and (38) is shown in Figure 3. The figure shows that the approximate results agree well with those of Eq. (37) except for large  $\varepsilon$  in the case of  $2(\gamma - \alpha I_s) < \alpha^2 \sigma^2$ . As the noise increases, the correlation time rapidly decreases, but the noise has no effect on the stability of the system.

Using the following assumption based on Eq. (29), we obtain

$$\langle x(t)^4 \rangle = \langle x(t)^2 \rangle [\langle x(t)^2 \rangle + \alpha^2 \sigma^2],$$
  
$$\langle x(t)^3 x(0) \rangle = [\langle x(t)^2 \rangle + \alpha^2 \sigma^2] G(t),$$
 (39)

The variance and correlation function at  $X_s = 0$  are given by

$$-\frac{d}{dt} \langle x(t)^2 \rangle = 2\{\gamma - \alpha I_s + \alpha^2 \sigma^2 \varepsilon^2 (\gamma - \alpha I_s + 4\alpha^2 \sigma^2)\} \langle x(t)^2 \rangle$$
$$-2(1 - 4\varepsilon^2 \alpha^2 \sigma^2) \langle x(t)^2 \rangle^2. \tag{40a}$$

$$-\frac{d}{dt}G(t) = \left[\gamma - \alpha I_s - \langle x(t)^2 \rangle - \alpha^2 \sigma^2\right]G(t). \tag{40b}$$

The time-dependent variance satisfies

$$\frac{\langle x(t)^2 \rangle}{\langle x(0)^2 \rangle} \left[ \frac{a' - b' \langle x(0)^2 \rangle b'}{a' - b' \langle x(t)^2 \rangle} \right] = \exp 2a't \tag{41}$$

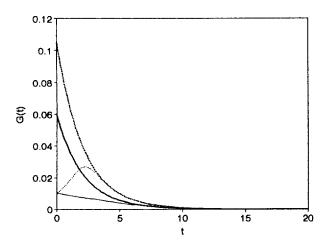
where

$$a' = \gamma - \alpha I_s + \varepsilon^2 \alpha^2 \sigma^2 (\gamma - \alpha I_s + 4\alpha^2 \sigma^2), b' = 1 - 4\varepsilon^2 \alpha^2 \sigma^2.$$
 (42)

In the  $\varepsilon \rightarrow 0$  limit we may rewrite Eq. (41) as

$$\langle x(t)^2 \rangle = \frac{\langle x(0)^2 \rangle (\gamma - \alpha I_s) \exp 2(\gamma - \alpha I_s)t}{\gamma - \alpha I_s + \langle x(0)^2 \rangle [\exp 2(\gamma - \alpha I_s)t - 1]}.$$
 (43)

Figure 4 shows that for small  $\varepsilon$  the results given in Eq.



**Figure. 5.** The comparison of correlation functions at the unstable steady state given in Eqs. (45) and (46). The heavy dotted and heavy solid lines denote the approximate results of Eq. (46) for  $(\gamma - \alpha I_s) > (\text{and } <) \alpha^2 \sigma^2$ , respectively. The dotted and solid lines are the results of Eq. (45) for  $(\gamma - \alpha I_s) > (\text{and } <) \alpha^2 \sigma^2$ , respectively. The values of parameters are  $\gamma = 2$ ,  $\alpha = 0.3$ ,  $\langle x(0)^2 \rangle = 0.01$  and  $\sigma = 2.2$ . The light intensity  $I_s = 3$  (and 6) is taken for  $\gamma - \alpha I_s > \alpha^2 \sigma^2$  ( $\gamma - \alpha I_s < \alpha^2 \sigma^2$ ).

(41) and (43) correspond well each other. For  $t\rightarrow\infty$ , the variance of Eq. (43) corresponds to that of the steady state given in Eq. (29), that is,

$$\lim_{t \to \infty} \langle x(t)^2 \rangle = \langle x^2 \rangle_s = \gamma - \alpha I_s. \tag{44}$$

Eq. (44) means that the assumption of Eq. (39) is quite reasonable after long time. Substituting Eq. (43) into Eq. (40b), we have

$$G(t) = \langle x(0)^2 \rangle \left[ \frac{\gamma - \alpha I_s}{\gamma - \alpha I_s + \langle x(0)^2 \rangle \left[ \exp 2(\gamma - \alpha I_s)t - 1 \right]} \right]^{1/2}$$

$$\times \exp(\gamma - \alpha I_s - \alpha^2 \sigma^2)t. \tag{45}$$

After long time the correlation function reduces to

$$G(t) \simeq [\langle x(0)^2 \rangle (\gamma - \alpha I_s)]^{1/2} \exp{-\alpha^2 \sigma^2 t}.$$
 (46)

The correlation function of Eq. (46) is quite different from Eq. (10) because the linear term is offset by a nonlinear term and then the noise strength term remains only. The above result clearly shows that the noise stabilizes the unstable steady state. An example of the correlation function of Eqs. (45) and (46) is shown in Fig. 5. In the region of short time Eq. (46) are quite different from Eq. (45). After long time, however, the approximate result agrees well with that of Eq. (45).

#### Conclusions

We have studied the statistical properties of the Schlögl model with the first order transition when an external noise acts on the model. Let us summarize the main results of the present paper:

(1) The Fokker-Planck equation given in Eq. (18b) was obtained by the wide band perturbation method. Some re-

sults have been reported by using the different perturbation method, that is, the approximate Fokker-Planck operator or the approximate renormalized equation of evolution for the Gaussian white noise.<sup>13~18,23</sup> The results except that of Hanggi *et al.*<sup>16</sup> correspond to the present result for the long time when the transient effects are neglected.

- (2) If  $2(\gamma \alpha I_s) > \alpha^2 \sigma^2$ , the stationary probability distribution is a binodal Gaussian distribution with the maximal peaks at  $\pm [\gamma \alpha I_s \alpha^2 \sigma^2/2]^{1/2}$ . The peaks indicate that the deterministic stable steady states  $\pm [\gamma \alpha I_s]^{1/2}$  are shifted to  $\pm [\gamma \alpha I_s \alpha^2 \sigma^2/2]^{1/2}$  by the noise strength in the  $\epsilon \to \infty$  limit. When  $2(\gamma \alpha I_s) = \alpha^2 \sigma^2$ , the distribution becomes Gaussian with the peak at x = 0. As the light intensity or the noise strength increases further, it becomes a delta-like distribution.
- (3) The noise strength decreases the correlation time between the fluctuating macroscopic variables when the system is at the stable steady state. It has no effect on the stability of the system. When the noise of very strong strength is applied to the system, the order of the system based on the deterministic equation is destroyed and thus the validity of the Fokker-Planck equation is a question to be solved.
- (4) The simple result of Eq. (45) directly shows that the external noise stabilizes the unstable steady state.

We have pointed out some results of the present work. Let us mention the important points which were not considered:

- (A) The external noise, which satisfies the non-Gaussian stochastic process, may give rise to phenomena which do not occur for Gaussian white noise. 10~11.24
- (B) When internal and external fluctuations act simultaneously on a system with small size, both effects on the nonequilibrium behaviors of the system have to be discussed together.

The present model will be extended by using the two points mentioned above.

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- 21. In the present model the photochemical reaction is assumed to occur at the third elementary step. We may also consider that the first step is the photochemical reaction step. In this case the effect of light intensity on stability is different from the present result.
- 22. Using that  $1-\varepsilon h(x) \cong \exp[-\varepsilon h(x)]$  for  $\varepsilon < 1$ , the probability distribution can be written as

$$P_{s}(x) \cong P_{os}(x) \exp\{-\varepsilon^{2} \left[3(\gamma - \alpha I_{S}) - 4x^{2} + \frac{1}{\alpha^{2}\sigma^{2}}(\gamma - \sigma I_{s} - x^{2})^{2}\right]\}.$$

With this result the detailed discussions for bifurcation phenomena are given in Refs. 13 and 15.

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