# EFFECT OF A SHOT-NOISE GENERATOR ON OSCILLATIONS IN A SALNIKOV MODEL OF AN EXOTHERMAL CHEMICAL REACTION*

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Our purpose is to study the impact of chaos-generators on the dynamics of nonlinear systems. As an example two simplified models of chemical reactions have been chosen with different coupling to the external noise. Numerical analysis together with analytic predictions for a stationary situation show possibility of noise induced periodicity.

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## 1. Introduction

Nonlinear dissipative systems exhibit natural strong sensitivity to the influence of external noise. It has been found both theoretically [1] and experimentally [2] that the effect of external noise may lead to interesting and nontrivial effects. Deterministically observed stationary states of the system may be shifted and the noise may create new states which have no deterministic analogue. Moreover, the noise can induce transitions between stochastic states [3] which may be further investigated in terms of "stochastic rate laws" [4].

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The effect of multiplicative noise in oscillatory systems has been considered in a great variety of physico-chemical examples [5]. In the case of a well understood electrical parametric oscillator [2], experimental results reported stabilization of the non-oscillatory regime in the presence of noise. The theoretical explanation of these findings has been based on the detailed analysis of the Hopf bifurcation by use of a reduced system of equations leading to a universal normal form [6]:

$$\frac{dz}{dt} = (a + i\Omega)z - Kz|z|^2 , \tag{1.1}$$

where $z$ is a complex amplitude and $a$, $\Omega$ and $K$ are real fluctuating parameters. Exact solutions to the Fokker–Planck equation describing the dynamics of fluctuations near instability has been obtained for a class of models [5] by imposing white noise perturbations on the model parameters $a$, $\Omega$, $K$.

The goal of this paper is to study a similar type of a noise-induced behaviour in "real" oscillatory system in which $a$ and $\Omega$ are complex dimensionless combinations of physical and chemical parameters describing the process of combustion.

We do not limit ourselves to a gaussian white noise idealization of fluctuations in the parameters. Instead, we investigate dynamic properties of the system perturbed by a more realistic shot-noise generator, whose statistical properties and longtime behaviour are well known [7], [8].

In Section 2 we briefly present the model and discuss stationary properties of fluctuations imposed on the system.

The simulation results are represented and analyzed in Section 3. Some comparative analysis of the dynamics with a more primitive model of a chemical reaction exhibiting oscillatory states (Brusselator) is discussed.

Finally, concluding remarks are found in Section 4.

## 2. The model

The model consists of two consecutive first-order chemical reactions

$$A \xrightarrow{k_0} X \xrightarrow{k_1(T)} B + Q , \tag{2.1}$$

converting an initial reactant $A$ into a final product $B$ through a single intermediate species $X$. The second reaction is exothermic and the rate constant $k_1(T)$ obeys an Arrhenius temperature dependence

$$k_1(T) = k_1^0 \exp\left( - \frac{E}{RT}\right) , \tag{2.2}$$

where $E$ is the activation energy. For simplicity we have assumed that the first reaction is thermoneutral and has zero activation energy ($k_0$ does not vary with temperature); as for the energy transfer we have assumed a Newtonian cooling (*i.e.*, proportional to the temperature difference).

Dimensionless form of mass and energy balance for (2.1) leads to a set of equations [9]:

$$\frac{d}{dt}\alpha = \mu - \kappa\alpha \exp\left(\frac{\theta}{1+\gamma\theta}\right),$$
$$\frac{d}{dt}\theta = \alpha \exp\left(\frac{\theta}{1+\gamma\theta}\right) - \theta, \qquad (2.3)$$

where $\theta$ stands for the dimensionless temperature rise, $\alpha$ represents dimensionless concentration of $X$ and $\mu$, $\kappa$, $\gamma$ are parameters. In particular $\gamma$ represents the ratio $\gamma = RT_\alpha/E$, where $R$ is gas constant, $T_\alpha$ is ambient temperature and $E$ — activation energy. A typical value of $\gamma$, being a dimensionless measure of the activation energy, is less then 1.

The model can display oscillatory behaviour and the conditions for the Hopf bifurcation require [6,10]:

$$\kappa^* = \left(\frac{\theta^*_{ss}}{(1+\gamma\theta^*_{ss})^2 - 1}\right) \exp\left(-\frac{\theta^*_{ss}}{(1+\gamma\theta^*_{ss})}\right),$$
$$\mu^* = \kappa^*\theta^*_{ss}, \qquad (2.4)$$

where $\theta^*_{ss}$ is a stationary-state temperature rise.
Direct analysis of (2.4) shows that Hopf bifurcation cannot occur if the activation energy $E$ becomes too small in comparison with the thermal energy $RT_\alpha$, *i.e.* if $E < 4RT_\alpha$ or equivalently if $\gamma > \frac{1}{4}$ (*cf.* Fig. 1).

Close to the bifurcation point the system (2.3) can be linearized to the form:

$$\begin{pmatrix}\dot{x}\\ \dot{y}\end{pmatrix} = \begin{pmatrix}a & -\Omega\\ \Omega & a\end{pmatrix}\begin{pmatrix}x\\ y\end{pmatrix} + \begin{pmatrix}f(x,y)\\ g(x,y)\end{pmatrix}, \qquad (2.5)$$

where $x, y$ measure excess of $\alpha, \theta$, from their stationary values $\alpha_{ss}, \theta_{ss}$. The system can be easily transformed to the form (1.1). A real part of the complex coefficient $K$ can be expressed in terms of partial derivatives of functions $f, g$:

$$\mathrm{Re}\,K = \frac{1}{16}\Big((f_{xxx} + f_{xyy} + g_{xxy} + g_{yyy})$$
$$+\frac{1}{\Omega}\big(f_{xy}(f_{xx} + f_{yy}) - g_{xy}(g_{xx} + g_{yy}) - f_{xx}g_{xx} + f_{yy}g_{yy}\big)\Big). \qquad (2.6)$$

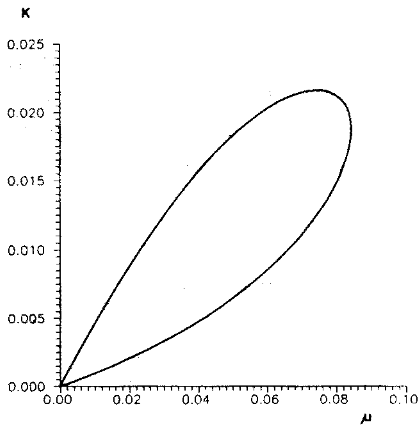


**Fig. 1. Positions of Hopf bifurcation points of the model (2.3) in $\kappa$ — $\mu$ parameter plane for $\gamma = 0.21$. Outside the curve stationary solution for any parameter values is a stable focus, inside — a stationary state is unstable and is surrounded by a stable limit cycle.**

According to the Hopf bifurcation theorem [6], periodic solutions of (2.3) parameterized by $\mu, \kappa$ and $\gamma$ are stable limit cycles if $\mathrm{Re}\, K < 0$ and are repelling if $\mathrm{Re}\, K > 0$.

With the full Arrhenius dependence, the model yields [11] the following form of the stability parameter $\mathrm{Re}\, K$:

$$\mathrm{Re}\, K = \frac{1 - \theta^*_{ss} - 2\gamma(3 - 2\theta^*_{ss}) - 3\gamma^2\theta^*_{ss}(4 - \theta^*_{ss}) - 6\gamma^3\theta^{*2}_{ss}}{8(1 + \gamma\theta^*_{ss})^6} \tag{2.7}$$

From the above expression it becomes readily understood that some combinations of the dimensionless activation energy and the stationary-state temperature rise may change the stability of the limit cycle emerging in the system *via* the Hopf bifurcation.

In the following we examine the behaviour of the system when the parameter $\gamma$ fluctuates according to regular perturbations $y(t)$ imposed on its average:

$$\gamma(t) = \gamma_0 + \sigma y(t) \tag{2.8}$$

and

$$\dot{y}(t) = -\tau^{-1}_{\mathrm{corr}} y(t) + \xi\,,$$
$$\xi = \tau^{1/2} \sum_n^{\infty} x_n \delta(t - n\tau)\,, \tag{2.9}$$

$\tau_{\mathrm{corr}}$ defines an intrinsic relaxation time of $y$, $\tau^{1/2} x_n$ stands for the intensity of "kicks" $\xi$ whose distribution is assumed to be generated by a logistic map:

$$x_{n+1} = \mathrm{T} x_n = 2x_n^2 - 1\,. \tag{2.10}$$

For $\tau \to 0$, Eqs (2.9) (2.10) are known [12,13] to be equivalent to the Langevin equation, provided initial $x_0$ is distributed according to some smooth probability distribution and T has so called $\varphi$-mixing property (in particular, $\varphi$-mixing property is inherent for the mapping (2.10)). The process $y(t)$ converges then to the continuous time Ornstein–Uhlenbeck process in velocities (and $\gamma(t)$ represents an "integrated" O-U process in positions).

Statistical properties of $y(t)$ can be easily verified [13] by use of a "propagator" $\lambda$:[1]

$$y_n = \lambda^n y_0 + \tau^{1/2} \sum_{l=0}^{n-1} \lambda^{n-1-l} x_l ,$$
$$\lambda = \exp\left( - \frac{\tau}{\tau_{\mathrm{corr}}} \right) \tag{2.11}$$

and integration is done between subsequent time steps $n\tau < t < (n+1)\tau$ (for extended analysis of the process (2.9), see *e.g.*[12]).

The first moments and correlation functions of $x_n$ can be derived straightforward basing on the ergodic properties of the map (2.10) [7,8]:

$$\langle x_n \rangle = 0 ,$$
$$\langle x_{n_1}, x_{n_2} \rangle = \tfrac{1}{2} \delta(n_1, n_2) ,$$
$$\langle x_{n_1}, x_{n_2}, x_{n_3} \rangle = \tfrac{1}{8} \sum_{\mathrm{P}(i_1,i_2,i_3)} \delta\left(n_{i_1}, n_{i_2} + 1\right) \delta\left(n_{i_1,i_3} + 1\right) , \tag{2.12}$$

where $\sum_{\mathrm{P}(i_1,i_2,i_3)}$ is the sum taken over 3! permutations of indices.

From (2.11) and (2.12) we get following expressions for the averages:

$$\langle y_n^2 \rangle = \lambda^n y_0 ,$$
$$\langle y_n^2 \rangle = \left(\lambda^n y_0\right)^2 + \tfrac{1}{2}\tau \frac{1-\lambda^{2n}}{1-\lambda^2} ,$$
$$\langle y_n^3 \rangle = \left(\lambda^n y_0\right)^3 + \tfrac{3}{4}\tau^{3/2} \frac{1-\lambda^{3n}}{1-\lambda^3} + \tfrac{3}{2}\tau \frac{1-\lambda^{2n}}{1-\lambda^2} \lambda^n y_0 \tag{2.13}$$

which in the long time limit $(n \to \infty)$ tend to:

$$\langle y_n^2 \rangle \to 0 ,$$
$$\langle y_n^2 \rangle \to \tfrac{1}{2} \frac{\tau}{1-\lambda^2} = \tfrac{1}{2} \frac{\tau}{1 - \exp\left(-2\tau/\tau_{\mathrm{corr}}\right)} ,$$
$$\langle y_n^3 \rangle \to \tfrac{3}{4}\tau^{3/2} \frac{\lambda^2}{1-\lambda^3} = \tfrac{3}{4}\tau^{3/2} \frac{\exp\left(-\frac{2\tau}{\tau_{\mathrm{corr}}}\right)}{1-\exp\left(-\frac{3\tau}{\tau_{\mathrm{corr}}}\right)} . \tag{2.14}$$

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[1] By integrating (2.9) with (2.10) one gets the recurrence relation used in our numerical analysis pf the noise: $y_{n+1} = \lambda y_n + \tau^{1/2} x_n$.

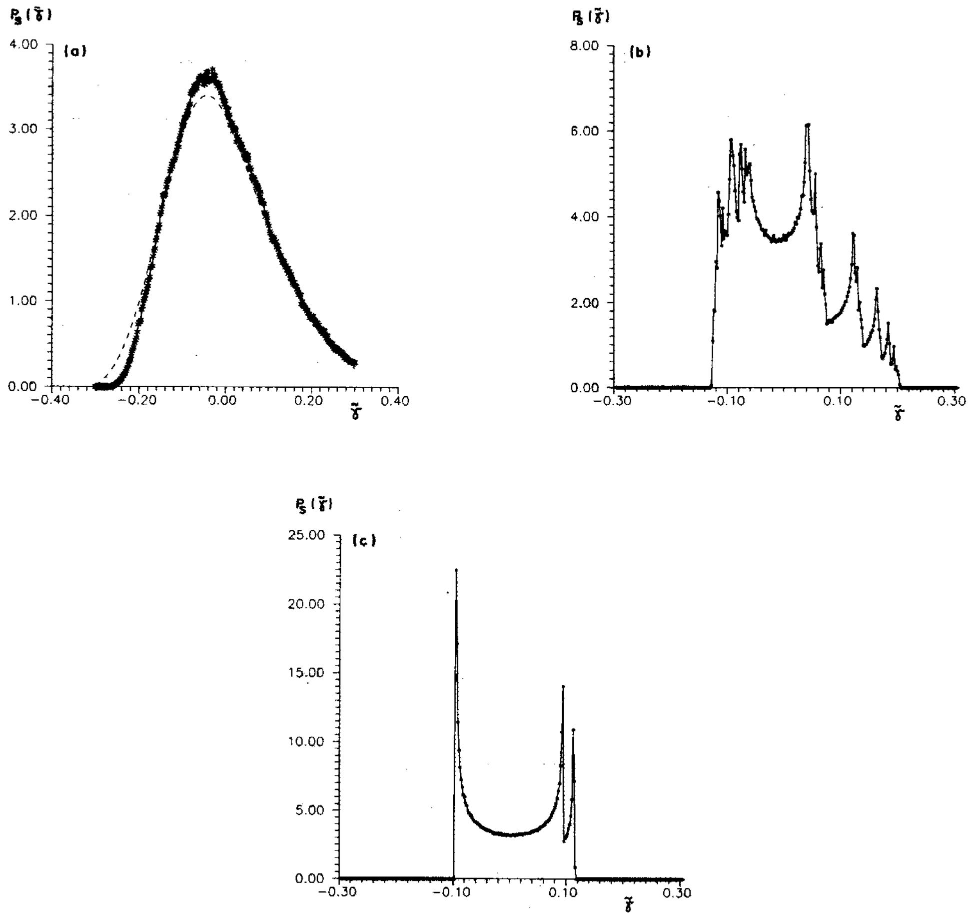


**Fig. 2. Stationary probability distribution of $\tilde{\gamma} = \gamma(t) - \gamma = \sigma y(t)$. Parameter values: (a) — $\lambda = 0.8$, $\sigma = 0.1$, $\tau = 1$, (b) — $\lambda = 0.5$, $\sigma = 0.1$, $\tau = 1$, (c) — $\lambda = 0.1$, $\sigma = 0.1$, $\tau = 1$. Dashed line stands for function (2.16).**

Asymptotically, stationary probability distribution of $y$, $P_s(y)$, can be approximated by a Gaussian and deviation from it can be represented by the Gram–Charlier expansion [14]:

$$P_s(y) = \frac{\exp\left(-\frac{y^2}{2\sigma^2}\right)}{\sigma(2\pi)^{1/2}}\left(1 + \sum_{k=0}^{\infty} c_k H_k(y)\right). \tag{2.15}$$

The functions $H_k$ are Hermite polynomials, $\sigma$ stands for the dispersion of $y$, and the coefficients $c_k$ are determined by the moments $\langle(\delta y)^n\rangle$, $\delta y = y - \langle y\rangle$.

Direct use of (2.14) yields:

$$P_s(y) = \left(\frac{1-\lambda^2}{\pi}\right)^{1/2} \exp\left(-y^2(1-\lambda^2)\right) * \left(1 + \frac{\lambda^2}{1-\lambda^3}\left(y^3(1-\lambda^2)^3 - \tfrac{3}{2}(1-\lambda^2)^2 y\right) + \tfrac{1}{24}\left(\frac{4}{1+\lambda^2} - 3\right)\left(4y^4(1-\lambda^2)^2 - 12(1-\lambda^2)y^2 + 3\right)\right). \quad (2.16)$$

Figures 2 (a), (b), (c) present stationary probability distributions of $\tilde{\gamma} = \gamma(t) - \gamma_0 = \sigma y(t)$ affected by fluctuations $y(t)$ with distinct parameters $\tau_{\mathrm{corr}}$ (*cf.* (2.11)). One can see that for $\lambda \simeq 1$ formula (2.16) gives quite satisfactory results, whereas for $\lambda \to 0$ the distribution does not remain gaussian.

## 3. Impact of a shot-noise generator on the dynamics of a Salnikov model

Typical behaviour of the model and its dynamic properties viewed in functional dependence of the parameters $\mu$, $\kappa$ have been discussed elsewhere [10], [15]. Our aim is to check evolution of the system in the domains of deterministic determined regions os steady states (*cf.* Fig. 1)

For a value of $\gamma_0 = 0.21$ (chosen to fulfill the Hopf bifurcation criterion: $\gamma_0 < 0.25$), and $\mu = 0.04$, $\kappa = 0.015$ (the conditions sufficient to get deterministic, stable limit cycle oscillations), we have operated with numerically generated noise $\{y(t)\}$ by varying values of $\sigma$ and $\tau_{\mathrm{corr}}$ (*cf.* Figs 3, 4).

The trajectories of the system (2.3), (2.8), (2.9) have been generated in the iteration of about $10^5$ steps. At the same correlation time of the noise $\tau_{\mathrm{corr}}$ and by varying intensity of the noise $\sigma$, we have observed emergence of a stochastic driven stable limit cycle with the amplitude increasing with $\sigma$.

Due to complexity of the system (nonlinearity of Eq. (2.3) does not allow for a standard evaluation of a respective normal form close to the bifurcation point in the presence of noise), one is not able to determine parametrically conditions for occurrence of a "stochastic Hopf bifurcation". In fact, exponential form of the kinetic equations would require an infinite series of moments used in the elimination of noise variable. We are assuming therefore that the system is exposed to a longtime limit form of the noise whose stationary properties are then well known [13].

In particular, for a stationary noise $\{y(t)\}$ we get:

$$\begin{aligned} \langle\gamma\rangle &= \gamma_0 , \\ \langle\gamma^2\rangle &= \gamma_0^2 + \frac{\sigma^2}{2}\frac{\tau}{1-\exp\left(-\frac{2\tau}{\tau_{\rm corr}}\right)} , \\ \langle\gamma^3\rangle &= \gamma_0^3 + \frac{3}{4}\sigma^3\tau^{3/2}\frac{\exp\left(-\frac{2\tau}{\tau_{\rm corr}}\right)}{1-\exp\left(-\frac{3\tau}{\tau_{\rm corr}}\right)} \\ &\quad + \frac{3}{2}\sigma^2\gamma_0\tau\frac{1}{1-\exp\left(-\frac{2\tau}{\tau_{\rm corr}}\right)} . \end{aligned} \tag{3.1}$$

For the parameters chosen

$$\gamma_0 = 0.21 \text{ and } \kappa = 0.015 < \kappa^*_{\rm max} = (1-4\gamma)e^{-2} = 0.022 ,$$

which is the maximum value for Hopf bifurcation to occur at a given $\gamma_0$ parameter, deterministic steady state is a stable limit cycle which looses stability along the upper branch of Hopf bifurcation points[2] (bifurcation degenerates). In fact, from (2.7) one gets change of the stability of the limit cycle at the value $\theta^{**}_{\rm ss} = 9.344$ (which is the upper branch of Hopf bifurcation points) if at the same time $\kappa$ becomes smaller than the corresponding $\kappa^*$ obtained from Eq. (2.4), *i.e.* $\kappa < \kappa^* = 2.767{}^*10^{-3}$. The lower branch of Hopf bifurcation points consists (for commonly chosen parameters) of nonphysical states ($\theta < 0$).

In the presence of noise, the determinant of the quadratic equation for $\theta^{**}_{\rm ss}$ (see numerator in Equation (2.7)) depends on averages $\langle\gamma\rangle$, $\langle\gamma^2\rangle$, $\langle\gamma^3\rangle$. For the set of parameters, $\gamma_0 = 0.21$, $\kappa = 0.015$, $\mu = 0.040$, after imposition of the noise, we observe appearance of a limit cycle shifted in the phase space as compared to the position of its deterministic analogue (*cf.* Fig. 3). The corresponding value of $\theta^{**}_{\rm ss}$, at which $\mathrm{Re}\, K$ changes the sign, can be calculated by use of (3.1) and reads $\theta^{**}_{\rm ss} = 22.3$, *i.e.* the noise stabilizes the position of a deterministic Hopf bifurcation point.

Figs 4 (a), (b) present a different situation where before the noise has been imposed on $\gamma$, the system possessed a single stationary state, *i.e.* a stable focus (now we have chosen $\gamma_0 = 0.21$, $\kappa = 0.015$, $\mu = 0.035$). Noise generated in $\gamma$ is shifting again the phase diagram of the system, so that one observes a limit cycle induced by external noise at, effectively, lower values of $\mu$.

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[2] Upper and lower branches of Hopf bifurcation points can be derived from (2.1) as the only solution for which $\kappa = 0$.

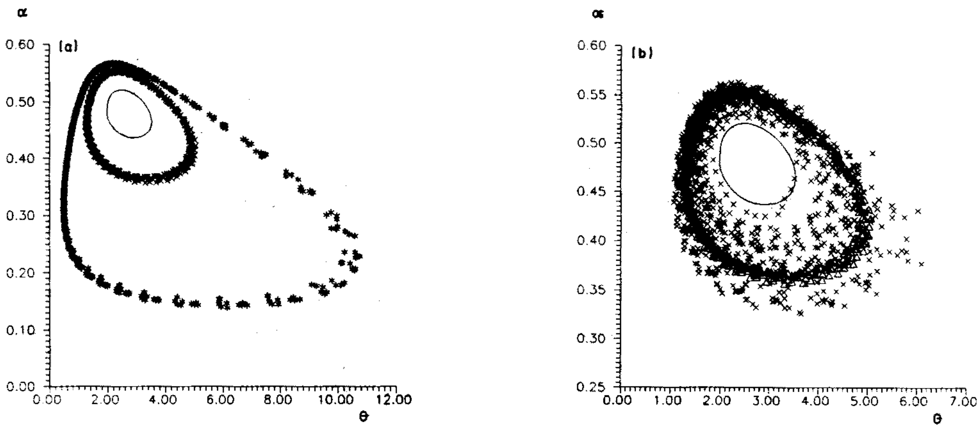


**Fig. 3. Phase portrait of the system (2.3) in the region of limit cycle oscillations ($\mu = 0.040$, $\kappa = 0.015$, $\gamma_0 = 0.21$) in the deterministic and stochastic case. (a) —— — deterministic LC, × — $\lambda = 0.1$, $\sigma = 0.05$, $\tau = 1$, $*$ — $\lambda = 0.1$, $\sigma = 0.1$, $\tau = 1$. (b) —— — deterministic LC, △ — $\lambda = 0.1$, $\sigma = 0.05$, $\tau = 1$, × — $\lambda = 0.8$, $\sigma = 0.05$, $\tau = 1$.**

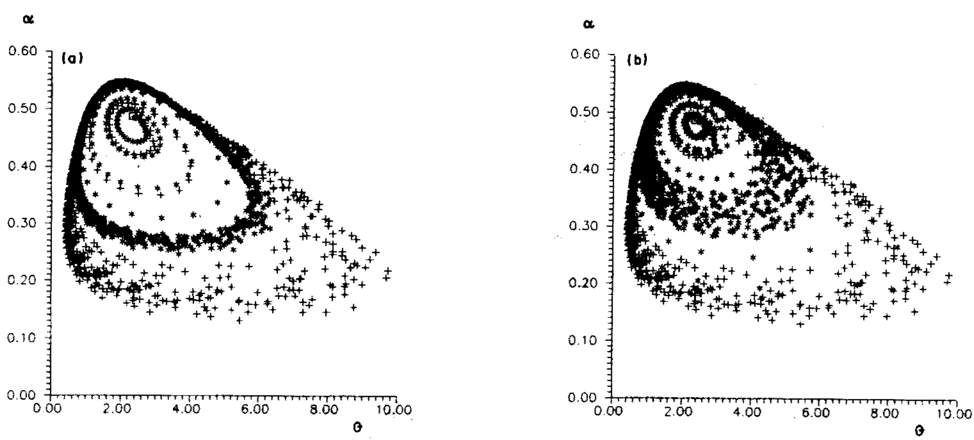


**Fig. 4. Phase portrait of the system (2.3) in the region of one stable stationary state ($\mu = 0.035$, $\kappa = 0.015$, $\gamma_0 = 0.21$) in the deterministic and stochastic case. (a) □ — deterministic stationary state, + — $\lambda = 0.5$, $\sigma = 0.1$, $\tau = 1$, $*$ — $\lambda = 0$, $\sigma = 0.1$, $\tau = 1$. (b) □ — deterministic stationary state, + — $\lambda = 0.5$, $\sigma = 0.1$, $\tau = 1$, $*$ — $\lambda = 0.5$, $\sigma = 0.08$, $\tau = 1$.**

This qualitatively new characteristic of the system is due to the very coupling of the dynamics (2.3) to the external noise in $\gamma$. It is only because of the highly nonlinear form of the evolution equation $(\alpha(t),\ \theta(t))$ that one observes occurrence of qualitatively different steady states in the region where, deterministic, their emergence would be forbidden. This behaviour is by no means a general phenomenon; instead, it is fully generic, depending on a particular form of a nonlinear evolution equation and the form of a coupling to the noise.

As a counter-example let us use the same type of noise to check stationary behaviour of a less complex model of a chemical reaction, *i.e.* the Brusselator, whose dynamics is also known to produce limit cycle oscillations. The model is described by the set of equations:

$$\begin{aligned} \dot{X} &= A - X + X^2Y - BX\,, \\ \dot{Y} &= BX - X^2Y\,. \end{aligned} \tag{3.2}$$

For the sake of clarity we chose $A = 1$. The system (3.2) is equivalent to:

$$\begin{aligned} \dot{x} &= y + 2xy + x^2y - x^3\,, \\ \dot{y} &= -x\,, \end{aligned} \tag{3.3}$$

where $x$ and $y$ determine variations from steady state $(X_s, Y_s) = (A, B/A) \equiv (1,1)$ known to undergo Hopf bifurcation for $B_c = 1+A^2 \equiv 2$. Let us assume further that $B$ fluctuates, $B = B_c + \gamma \equiv 2 + \gamma$, where $\gamma$ is described by (2.8), (2.9) with $\gamma_0 = 0.3$ (3.3) reads then:

$$\begin{aligned} \dot{x} &= y + 2xy + x^2y - x^3\,, \\ \dot{y} &= -x\,. \end{aligned} \tag{3.4}$$

To analyze behaviour of the system close to the Hopf bifurcation point, we restrict ourselves to the flow of (3.4) within the center manifold. By use of a smooth coordinate to produce eventually Eq. (1.1) with:

$$\begin{aligned} a &= \frac{\gamma}{2}\,, \\ \Omega &= \left(1 - \frac{\gamma^2}{4}\right)^{1/2}, \\ \mathrm{Re}\,K &= -\frac{3}{8} - \frac{7}{72}\gamma\,. \end{aligned} \tag{3.5}$$

As it can be easily seen from the form of $\mathrm{Re}\,K$, stationary (longtime limit) fluctuations of $\gamma(t)$ are not going to change stability properties of the emerging limit cycle provided intensity of perturbations is not too big to change the character of the bifurcation point ($\gamma^2$ has to be smaller than 4).

Fig. 5 presents results of the simulation of (3.3) with the noise of the form (2.9) imposed on the parameter $B = B_c + \gamma$. Obviously, this time the noise stabilizes the limit cycle observed in the purely deterministic case.

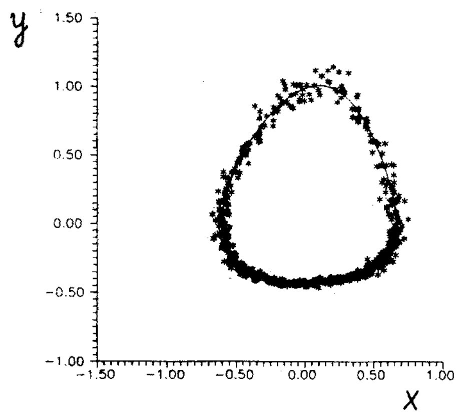


Fig. 5. Limit cycle oscillations in the Brusselator model; deterministic — ——, stochastic simulations — *. Parameter values: $A = 1$, $\gamma_0 = 0.3$, $B = 2.3$.

## 6. Conclusions

The concept of noise-induced transitions has achieved wide interest and has found numerous applications in theoretical studies of the effect of external noise in nonlinear systems. On the other hand, domains of chaotic motions embedded in mechanical processes driving time-evolution of a physical system are known to influence its dynamic properties [13], [7] in the similar way as it is observed in the presence of noise. This rises the question when and under what conditions chaotic motions can be source of a random behaviour and wether that could imply existence of a unified theoretical description in terms of mechanics and the theory of stochastic processes.

Theoretical and numerical investigations presented in this paper reveal richness of behaviours which so far remain unexplored and would require further experimental justifications. Noise, understood as a limit of a purely deterministic dynamic process (with the inherent chaotic regimes) is not simply a source of a disorder in a nonequilibrium system. Its presence may induce (even at relatively low intensities) quite organized behaviours and transition phenomena.

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