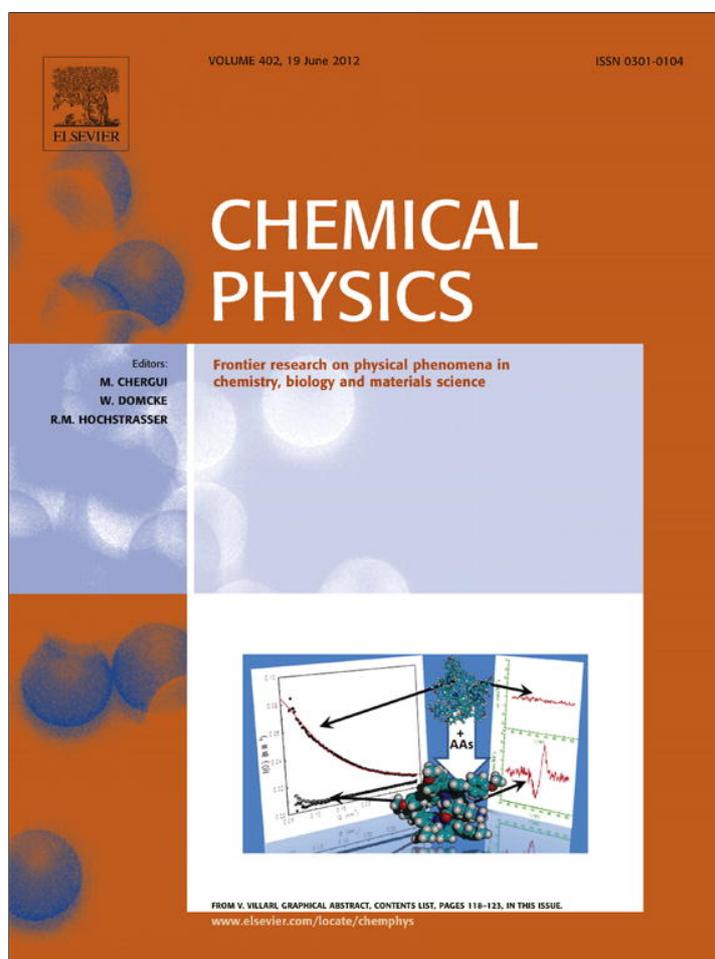


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Noise-and delay-induced phase transitions of the dimer–monomer surface reaction model

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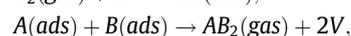
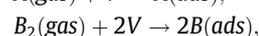
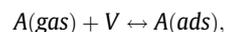
ABSTRACT

The effects of noise and time-delayed feedback in the dimer–monomer (DM) surface reaction model are investigated. Applying small delay approximation, we construct a stochastic delayed differential equation and its Fokker–Planck equation to describe the state evolution of the DM reaction model. We show that the noise can only induce first-order irreversible phase transition (IPT) characteristic of the DM model, however the combination of the noise and time-delayed feedback can simultaneously induce first- and second-order IPT characteristics of the DM model. Therefore, it is shown that the well-known first- and second-order IPT characteristics of the DM model may be viewed as noise-and delay-induced phase transitions.

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1. Introduction

The subject of kinetic or nonequilibrium phase transitions in surface catalysis is of great interest in physics, chemistry, biology, sociology, and even economics [1–4]. A number of nonequilibrium models such as DM [5–8], monomer–monomer [9–11], dimer–dimer [12,13], monomer–monomer–monomer [14,15], etc. exhibiting phase transitions, have been studied. Among all these lattice models used for describing nonequilibrium phase transitions of surface catalysed reactions, the DM reaction model: $A + \frac{1}{2}B_2 \rightarrow AB$, which is also known as the ZGB model [5]. This model is the archetypical one, it is based upon the Langmuir–Hinshelwood mechanism, i.e. both the reactants are adsorbed on the surface. It is assumed that the reaction occurs according to the following steps [16]:



where “V” denotes a vacant adsorption site, “gas” and “ads” refer to gaseous and adsorbed molecules or atoms, respectively. In a Monte Carlo simulation (MCS), the monomer A adsorbs at single empty sites with rate Y_A , the dimer B_2 adsorbs at adjacent pairs of empty sites with rate $Y_B = 1 - Y_A$, and the reaction between different

species adsorbed at adjacent pairs of sites occurs instantaneously. The most distinctive feature of the DM model is the occurrence of a first-order A-poisoning IPT at high $Y_A = Y_2$ and a second-order B-poisoning IPT at low $Y_A = Y_1$. It is found by computer simulation that below a critical value $Y_1 = 0.389 \pm 0.005$, the lattice becomes completely covered with B atoms and all reactions cease (the B-poisoning phase). Above a second value $Y_2 = 0.525 \pm 0.001$, the system enters the A-poisoning phase in which the lattice is completely covered with A molecule. In the intermediate case, $Y_1 < Y_A < Y_2$ (the reaction window), the system reaches a reactive steady state in which the reactions between A and B can proceed indefinitely.

There have been many theoretical developments toward understanding such phenomena in the DM surface reaction model by mean field theory (MFT) [17–21]. Two typical types of approximations are used in MFT: one is the simple site approximation (SA) which neglects spatial correlation and assumes random distribution of sites. The other is the pair approximation (PA), which considers the nearest-neighbor correlation and assumes random distribution of pairs $i - j$ ($i, j = A, B, V$). Both SA-MFT and PA-MFT can qualitatively predict the phase transition behavior, but PA-MFT yields more correct quantitative predictions of the values of Y_{s1} and Y_{s2} , namely, one obtains $Y_{s1} = 0.2487$ and $Y_{s2} = 0.5241$ for PA-MFT and $Y_{s1} = 0$ and $Y_{s2} = 0.561$ for SA-MFT [17]. By comparison with the MCS values $Y_1 = 0.389$ and $Y_2 = 0.525$ [5], one sees that PA-MFT works quite well at Y_2 but not Y_1 , and the divergence between theory and simulation at Y_1 presumably indicates that long-range correlations should be considered near second-order phase transition [18,19]. Based on SA-MFT [17], another notes, however, the MCS value of the first-order IPT $Y_2 = 0.525$ is

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substantially below $Y_{s2} = 0.561$. In order to explain the discrepancy between Y_2 and Y_{s2} , Hou et al. considered the gas-phase fluctuations in the MCS [20], they obtained $Y_{s1} = 0$ and $Y_{s2} = 0.525$. The above investigations on the DM model, the better prediction of the first-order IPT-value Y_2 is obtained if the fluctuations of the adsorption rate Y_A is accounted for. In fact, the fluctuations of the control parameters can lead to external noise, which plays an important role in noise-induced transitions [22–24]. However, the second-order IPT-value Y_1 observed in simulation is poor. So, one devises that these investigations on the DM model may neglect possible effects induced by time-delayed feedback. In practice, in many physical as well as biological systems, time delays always exist and play a significant role in the dynamics, such as biophysiological controls [25], signal transmissions in biological and artificial neuronal networks [26], and laser dynamics in optical cavities [27,28], etc. Meanwhile, it appears that the combination of noise and time delay is ubiquitous in nature and often change fundamentally dynamics of the system [29–34]. From these above discussions, one may conclude that the noise and time-delayed feedback should be responsible for the occurrence of the discrepancy between theory and simulation. It is shown that the combinations of the noise and time-delayed feedback can induce first- and second-order IPT characteristics of the DM surface reaction model.

In this paper, we provide a theoretical analysis for the first- and second-order IPTs of the DM model if both the noise and time-delayed feedback are accounted for. In Section 2, we construct a stochastic delayed differential equation and the corresponding Fokker Planck equation, and then the effects of the noise and time-delayed feedback on the bifurcation diagram are discussed. Finally, concluding remarks are given in Section 3.

2. The model

We consider the DM surface reaction model (or ZGB model), the motions for the concentrations (y_A and y_B) are determined by the following two coupled ordinary differential equations [17]

$$\frac{dy_A}{dt} = Y_A y_V (1 - y_B)^4 - 2Y_B y_V^2 [1 - (1 - y_A)^3], \quad (1)$$

$$\frac{dy_B}{dt} = 2Y_B y_V^2 (1 - y_A)^3 - Y_A y_V [1 - (1 - y_B)^4], \quad (2)$$

where y_i ($i = A, B$ or V) denotes i -site concentration, and $y_V = 1 - y_A - y_B$. The first term in Eq. (1) represents the increase in y_A due to an A adsorbing on a vacant site with no B neighbors, and the second term represents the decrease in y_A due to a B adsorbing on a vacant site with at least one A neighbor and reacting. Eq. (2) is similarly constructed.

To obtain the Langevin equation and the corresponding Fokker-Planck equation, we will first reduce the two coupled ordinary differential equations to the state evolution equation of y_A , and then consider Y_A and y_A as the stochastic and time-delayed variables, respectively. Note that in the steady state, the sticking probability of A and B_2 are exactly equal [17]:

$$Y_A y_V - 2Y_B y_V^2 = 0. \quad (3)$$

Thus, one expects that the Eq. (3) also holds approximately near the steady state [20],

$$Y_A y_V - 2Y_B y_V^2 = \lambda, \quad (4)$$

in which λ is a small quantity near zero. Therefore one has

$$y_V \simeq \frac{Y_A}{2Y_B} - \frac{\lambda}{Y_A} = Y, \quad (5)$$

where $Y_B = 1 - Y_A$. Substituting Eq. (5) back into Eqs. (1) and (2), one has

$$\frac{dy_A}{dt} = Y_A Y [(1 - y_B)^4 + (1 - y_A)^3 - 1] - \lambda [(1 - y_A)^3 - 1], \quad (6)$$

$$\frac{dy_B}{dt} = Y_A Y [(1 - y_B)^4 + (1 - y_A)^3 - 1] - \lambda (1 - y_A)^3. \quad (7)$$

Notice that near the second-order B-poisoning IPT at low $Y_A = Y_1$ and first-order A-poisoning IPT at high $Y_A = Y_2$, i.e. dy_A/dt and dy_B/dt have contrasting signs. If Y_A is very close to Y_1 (or Y_2), the MCS shows that before the system is poisoned by A and B, or reaches the reactive steady state, there exists a long relaxation time when y_V hardly varies [5,17,20]. Therefore, the derivative of $y_A + y_B = 1 - y_V$ is

$$\frac{dy_A}{dt} + \frac{dy_B}{dt} \simeq 0. \quad (8)$$

Substituting Eq. (7) into Eq. (8), we obtain the deterministic equation of motion with respect to y_A ,

$$\frac{dy_A}{dt} = -Y_A Y [(Y + y_A)^4 + (1 - y_A)^3 - 1] + \lambda (1 - y_A)^3, \quad (9)$$

where Y is given by Eq. (5).

In most practically relevant cases, the state of the system can exhibit temporal long range correlations and should be affected in the first place by its immediate past, with additional correction arising from the time-delayed feedback [29,30]. Such an extension is important since it has been realized that time delays are ubiquitous in nature and often change fundamentally the dynamics of the system [31–34]. Namely, when the time-delayed feedback is added to the Eq. (9), the delayed differential equation of motion for the concentration is

$$\frac{dy_A}{dt} = -Y_A Y [(Y + y_A)^4 + (1 - y_A)^3 - 1] + \lambda (1 - y_A)^3 - \epsilon y_{A\tau}, \quad (10)$$

where $y_{A\tau}$ denotes the time-delayed variable $y_A(t - \tau)$, τ is the delay time, and ϵ is the intensity of the delayed feedback.

We now consider the fluctuation of Y_A by a stochastic parameter $Y_A + \eta(t)$, where $\eta(t)$ is the Gaussian white noise with zero mean, and the autocorrelation $\langle \eta(t)\eta(t') \rangle = 2\sigma\delta(t - t')$, where σ is the noise intensity. Then, Y_A in Eq. (10) is replaced by $Y_A + \eta(t)$, a linear expansion in powers of the noise $\eta(t)$ can be used to obtain a multiplicative noise term $\sim g(y_A, \lambda)\eta(t)$, where $g(y_A, \lambda)$ is the multiplicative function of the noise $\eta(t)$, it is given by (also see Ref. [20])

$$g(y_A, \lambda) = -\frac{Y_A(2 - Y_A)}{2(1 - Y_A)^2} [(Y + y_A)^4 + (1 - y_A)^3 - 1] + 4Y_A Y \left[\frac{1}{(1 - Y_A)^2} - \frac{\lambda}{Y_A^2} \right] (Y + y_A)^3. \quad (11)$$

Thus, the stochastic delayed differential equation under the Stratonovich form (or Langevin equation) corresponding to Eq. (10) reads (we rewrite y_A by y):

$$\frac{dy}{dt} = f(y, y_\tau, \lambda) + g(y, \lambda)\eta(t), \quad (12)$$

in which

$$f(y, y_\tau, \lambda) = -Y_A Y [(Y + y)^4 + (1 - y)^3 - 1] + \lambda (1 - y_A)^3 - \epsilon y_\tau, \\ g(y, \lambda) = -\frac{Y_A(2 - Y_A)}{2(1 - Y_A)^2} [(Y + y)^4 + (1 - y)^3 - 1] + 4Y_A Y \left[\frac{1}{(1 - Y_A)^2} - \frac{\lambda}{Y_A^2} \right] (Y + y)^3. \quad (13)$$

If $\sigma = 0$ and $\epsilon = 0$, the Eq. (12) with (13) degenerates to the original deterministic Eq. (9).

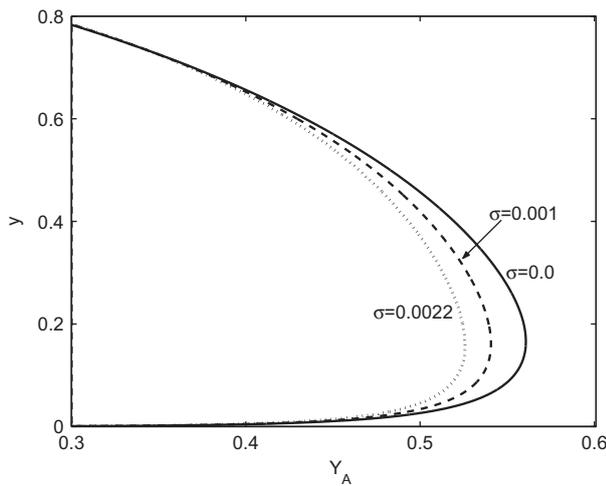


Fig. 1. Bifurcation diagram of the solutions of Eq. (20) as a function of Y_A for different values of σ .

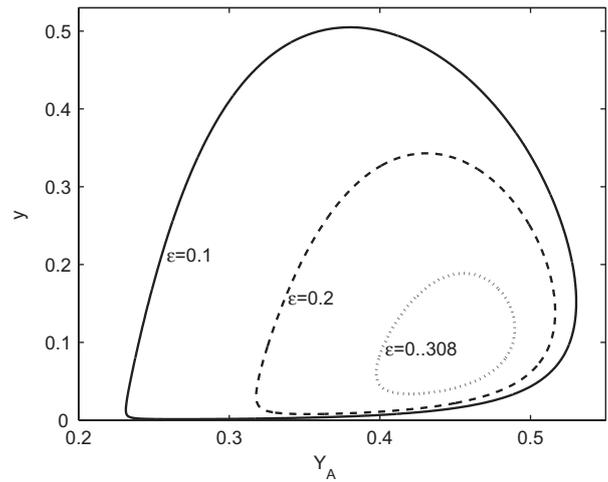


Fig. 2. Bifurcation diagram of the solutions of Eq. (19) as a function of Y_A for different values of ϵ with $\tau = 0.3$ and $\sigma = 0.001$.

Let $P(y, t)$ denotes the probability density distribution that the concentration exactly equals y at time t . Using the small delay approximation [31,32], the delayed Fokker–Planck equation of $P(y, t)$ corresponding to Eq. (12) can be given by

$$\frac{\partial P(y, t)}{\partial t} = -\frac{\partial}{\partial y} F(y, \lambda) P(y, t) + \frac{\partial^2}{\partial y^2} G(y, \lambda) P(y, t), \quad (14)$$

where the effective drift and diffusion coefficients $F(y, \lambda)$ and $G(y, \lambda)$ are described by

$$F(y, \lambda) = \sqrt{\frac{1}{2\pi D^{(0)}(y, \lambda)\tau}} \int_{-\infty}^{\infty} h(y, y_\tau, \lambda) \exp\left\{-\frac{[y_\tau - y - h^{(0)}(y, \lambda)\tau]^2}{2D^{(0)}(y, \lambda)\tau}\right\} dy_\tau, \quad (15)$$

$$G(y, \lambda) = \sqrt{\frac{1}{2\pi D^{(0)}(y, \lambda)\tau}} \int_{-\infty}^{\infty} k^2(y, \lambda) \exp\left\{-\frac{[y_\tau - y - h^{(0)}(y, \lambda)\tau]^2}{2D^{(0)}(y, \lambda)\tau}\right\} dy_\tau, \quad (15)$$

in which

$$h(y, y_\tau, \lambda) = f(y, y_\tau, \lambda) + \sigma g(y, \lambda) g'(y, \lambda), \quad (16)$$

$$k(y, \lambda) = \sqrt{\sigma} g(y, \lambda)$$

and $D^{(0)}(y, \lambda) = g^2(y, \lambda)$, $h^{(0)}(y, \lambda) = h(y, y_\tau, \lambda)|_{y_\tau=y}$. Substituting Eq. (16) into (15), we obtain

$$F(y, \lambda) = (1 - \epsilon\tau)(f^{(0)}(y, \lambda) + \sigma g(y, \lambda) g'(y, \lambda)), \quad (17)$$

$$G(y, \lambda) = \sigma g^2(y, \lambda),$$

where $f^{(0)}(y, \lambda) = f(y, y_\tau, \lambda)|_{y_\tau=y}$.

To obtain the steady state solution, one sets $\lambda = 0$ and $\partial P(y, t)/\partial t = 0$. Here, we would like to explain the role of λ used in the this paper. In practice, we first take advantage of λ to reduce the two coupled nonlinear equations to the evolution equation of y_A , Eq. (9), which is used to construct the stochastic delayed differential equation and corresponding Fokker–Planck equation. Finally, since we only consider the steady state bifurcation character of the system, we reset λ to zero. Thus, the stationary probability distribution (SPD) corresponding to Eq. (14) is given by

$$P_{st}(y, \lambda = 0) = \frac{N}{G(y, \lambda = 0)} \exp\left(\int^y \frac{F(x, \lambda = 0)}{G(x, \lambda = 0)} dx\right), \quad (18)$$

where N is a normalization constant. The effects of the noise and time-delayed feedback on the bifurcation diagram are given by

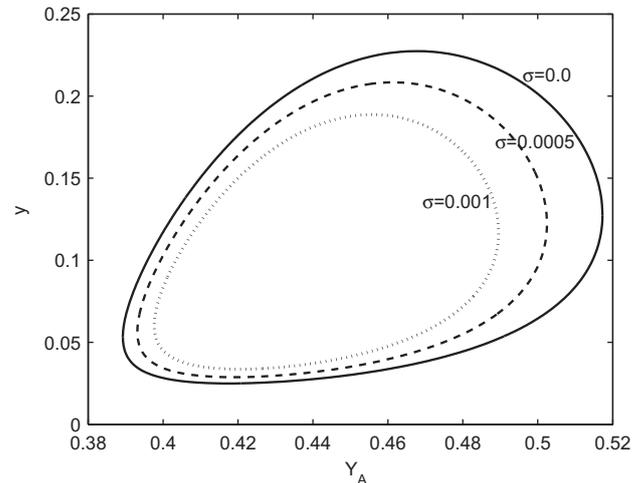


Fig. 3. Bifurcation diagram of the solutions of Eq. (19) as a function of Y_A for different values of σ with $\epsilon = 0.308$ and $\tau = 0.3$.

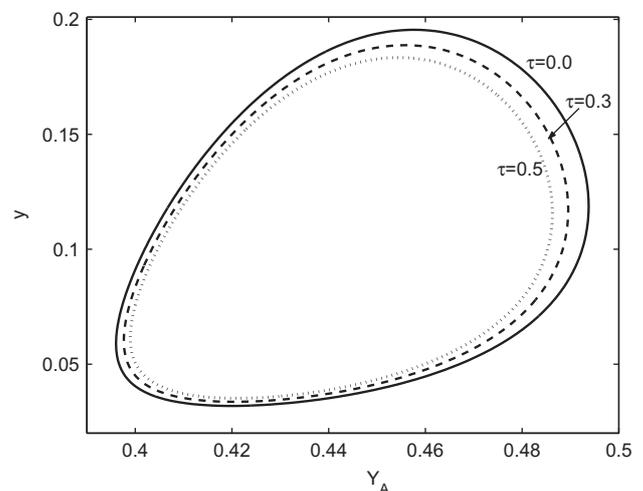


Fig. 4. Bifurcation diagram of the solutions of Eq. (19) as a function of Y_A for different values of τ with $\epsilon = 0.308$ and $\sigma = 0.001$.

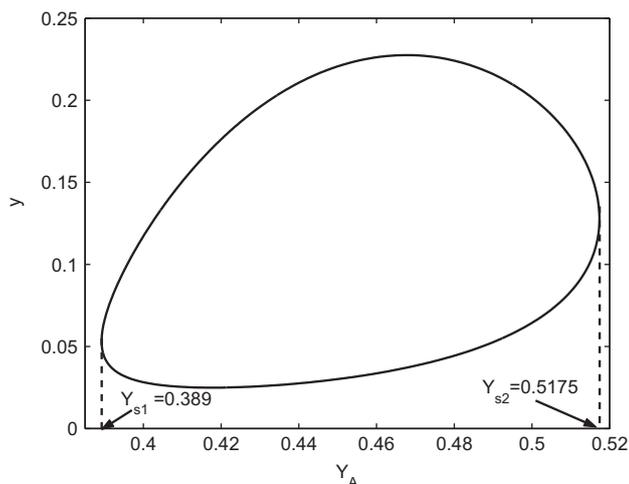


Fig. 5. Bifurcation diagram of the solutions of Eq. (19) as a function of Y_A with $\epsilon \simeq 0.308$, $\sigma = 0.0$ and $\tau = 0.0$.

the extrema of the SPD, the extrema of the SPD obeys a general equation $F(y, \lambda = 0) - G'(y, \lambda = 0) = 0$,

$$(1 - \epsilon\tau)f^{(0)}(y, \lambda = 0) - (1 + \epsilon\tau)\sigma g(y, \lambda = 0)g'(y, \lambda = 0) = 0. \quad (19)$$

For small values of Y_A , ϵ and τ , there are two solutions for Eq. (19), y_1 and y_2 ($y_1 \leq y_2$, where the lower branch y_1 is steady state and the higher branch y_2 unstable steady state), in the interval $[0, 1]$, which coalesce when $Y_A = Y_{s1}$ and $Y_A = Y_{s2}$, where Y_{s1} and Y_{s2} denote two spinodal points, respectively. If $\epsilon = 0$, one finds that the reactive steady state satisfies

$$f^{(0)}(y, \lambda = 0, \epsilon = 0) - \sigma g(y, \lambda = 0)g'(y, \lambda = 0) = 0, \quad (20)$$

so that a spinodal point exists at $Y_{s2} = 0.561$ if $\sigma = 0.0$, which is in agreement with Ref. [17] (or a spinodal point exists at $Y_{s2} \simeq 0.525$ in Ref. [20] if $\sigma \simeq 0.0022$).

The effects of the noise and time-delayed feedback on the bifurcation diagram are shown in Figs. 1–5. We first consider the effect of the noise on the bifurcation diagram. If $\sigma \neq 0$ and $\epsilon = 0$, as shown in Fig. 1. We show the solutions of Eq. (20) for different values of σ , and find only a left-shifting of the spinodal point at Y_{s2} as the value of σ increases. When $\sigma \simeq 0.0022$, the spinodal point exists at $Y_{s2} \simeq 0.525$, which coincides with the MCS value of the first-order IPT.

Next, we discuss the impacts of the combinations of the noise and time-delayed feedback on the bifurcation diagram. If $\epsilon \neq 0$ and $\sigma \neq 0$, we find two spinodal points at Y_{s1} and Y_{s2} . In Figs. 2–4, we show the solutions of Eq. (19) for different values of ϵ , σ and τ , respectively. We find that the right-shifting of the spinodal point at Y_{s1} and the left-shifting of the spinodal point at Y_{s2} as the values of ϵ , σ and τ increase, respectively. It is clear that the right-shifting of the spinodal point at Y_{s1} is more sensitive to the ϵ , and the left-shifting of the spinodal point at Y_{s2} is more sensitive to the σ and τ . For zero noise intensity $\sigma = 0.0$ and zero time delay $\tau = 0.0$, when $\epsilon \simeq 0.308$, as shown in Fig. 5. One sees that Y_{s1} increases from 0.0, which is the spinodal point of the deterministic equations of motion, to 0.389, which is obtained by MCS [5]. Another sees that Y_{s2} decreases from 0.561, which also is the spinodal point of the deterministic equations of motion, to 0.5175, which almost coincides with the MCS value [5].

3. Concluding remarks

In this paper, we have considered the fluctuation of Y_A and the time-delayed feedback of y_A in the DM surface reaction model, and constructed a stochastic delayed differential equation and its Fokker–Planck equation for the state evolution of the system. Using the small delay approximation, the SPD of the DM surface reaction model is obtained. The impacts of the noise and time-delayed feedback on the bifurcation diagram are given by the extrema of the SPD. If $\sigma \neq 0$ and $\epsilon = 0$, we only find a left-shifting of the spinodal point at Y_{s2} as the value of σ increases. When $\sigma \simeq 0.0022$, the spinodal point exists at $Y_{s2} \simeq 0.525$, which is in agreement with Ref. [20]. But if $\epsilon \neq 0$ and $\sigma \neq 0$, we find two spinodal points at Y_{s1} and Y_{s2} . It is shown that the right-shifting of the spinodal point at Y_{s1} and the left-shifting of the spinodal point at Y_{s2} as the values of ϵ , σ and τ increase, respectively, and the right-shifting of the spinodal point at Y_{s1} is more sensitive to the intensity ϵ of the delayed feedback, and the left-shifting of the spinodal point at Y_{s2} is more sensitive to the noise intensity σ and time delay τ . By “fine tuning” the time-delayed feedback and noise intensity (i.e., $\epsilon \simeq 0.308$, $\tau = 0.0$ and $\sigma = 0.0$), the two spinodal points exist at $Y_{s1} \simeq 0.389$ and $Y_{s2} \simeq 0.5175$, which coincides with the MCS of the first- and second-order IPTs, respectively [5]. Finally, we will discuss the impact of the small delay approximation on our results. For $\tau = 0.0$ and $\sigma = 0.0$, the Eq. (19) reduces as

$$-Y_A Y [(Y + y)^4 + (1 - y_A)^3 - 1] - \epsilon y = 0, \quad (21)$$

which is consistent with $dy_A/dt = 0$ of Eq. (10) for $\tau = 0.0$ and $\lambda = 0.0$. In others words, this set of $\tau = 0.0$ corresponds to that the Eq. (9) is modified by an additional linear term $-\epsilon y$ (see (10)). This new term produces the spinodal point at Y_{s1} which is absent in previous literature [17,20]. The delayed feedback ϵ in Eq. (21) plays crucial role in the vicinity of the first- and second-order IPTs (see Fig. 5), i.e., when $\epsilon \simeq 0.308$, we are able to obtain predictions in good agreement with simulation results. Therefore, the impact of the small delay approximation on our main results can be neglected.

In summary, the time-delayed feedback in the form of the difference between the current and a delayed state of the system can be used to control phase transitions that are induced by noise. It is shown that the well-known first- and second-order IPT characteristics of the DM reaction model may be viewed as noise- and delay-induced phase transitions. Although the noise and time-delayed feedback are important near the first- and second-order IPTs of the DM surface reaction model, we expect that the noise and time-delayed feedback also paly an important role in other surface reaction model, such as the monomer–monomer, dimer–dimer and monomer–monomer–monomer, etc.

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