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Novel effect of coupled external and internal noise in stochastic resonance[☆]

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Abstract

In this paper, on the basis of mesoscopic stochastic model of NO reduction by CO on single-crystal platinum surfaces, we report a novel effect of the external noise of reaction rate constant coupled to internal noise in stochastic resonance induced by external noise (SREN) or internal noise (SRIN). It is shown that the internal noise can enhance the SREN, and the external noise intensity for the SREN increases with increasing internal noise. However, the external noise can suppress the SRIN, and the suppressions nonmonotonously vary with the increasing external noise intensity. This result is different from the effect of the external noise of NO partial pressure coupled to the internal noise in the SR behavior of the system, which shows that the various external noise sources have different effects in the SRIN. And different roles of positive or negative feedback of the external noise terms may be a probable mechanism for the difference.

Keywords: Stochastic resonance; External noise; Internal noise; Chemical Langevin equation; Catalytic NO reduction by CO reaction

1. Introduction

Over the past two decades, stochastic resonance (SR) phenomena, a rather counterintuitive fact that the response of a nonlinear system to an external periodic signal may be enhanced through an optimal external noise, have been widely studied in physical, chemical, and biological systems [1–18]. In recent years, the effect of internal noise has attracted much attention, and the internal noise-induced SR phenomena have been observed in biological and chemical systems, including ion channel gating and neuron spiking [19–24], circadian clocks [25–28], intracellular calcium signaling [29,30], genetic regulation [31,32], CO oxidation on nanometer-sized particles or very small single-crystal surfaces [33–39], and NO reduction on small-size Pt surfaces [40,41]. Notably, the SR phenomena have already been observed in experiments [15–18,34–39]. It is shown that the internal noise can induce stochastic oscillations when the system is sub-threshold or supra-threshold, and the stochastic oscillations show the best performance at an optimal system size.

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It is known that the external noise may originate from the random variation of one or more of the externally set control parameters, such as reaction rate constants or partial pressures associated with a given set of reactions, while internal noise comes from the random fluctuations of the stochastic chemical reaction events [42–45] in a finite-size chemical system. Internal and external noise may simultaneously arise in finite-size chemical reaction systems, and hence they should be considered simultaneously.

In fact, the effects of external and internal noise on the oscillatory kinetics in biological and chemical reaction systems have already been studied. It was found that the external noise or the internal noise can either enhance or reduce the SR in ion channels [19–22]. External noise coherence resonance can be suppressed by internal noise, while internal noise coherence resonance can be enhanced by the modulation of external noise strength in a circadian oscillator [46]. However, the external and internal noise in these studies are usually considered independent, and this case is obviously unrealistic compared to the real systems in which the internal noise from the stochastic chemical reaction events might be associated with the external noise of reaction rate constants or gas partial pressures. Therefore, the external noise and internal noise are often coupled to each other, and this kind of coupling would cause different effects on the oscillatory kinetics of the systems. Recently, the effect of system size (internal noise) on the reaction oscillations and internal noise-induced SR have been studied [40,41]. Very recently, our study of the effect of the external noise of NO partial pressure coupled to internal noise in the system of NO catalytic reduction reaction has shown that the SR induced by the external noise (SREN) can be enhanced by the internal noise, and the SR induced by the internal noise (SRIN) can also be enhanced by the external noise [47]. Since the external noise may come from the random variations of various externally set control parameters, it is necessary and significant to investigate the effects of different kinds of external noises. The goal of this paper is to discuss how the coupled external noise of reaction rate constant and the internal noise affect the SREN and SRIN.

In this paper, based on the NO stochastic reaction model, we have investigated the effect of the external noise of reaction rate constant coupled to internal noise in the SREN and SRIN. It is found that an optimal internal noise can enhance the SREN, while the external noise can suppress the SRIN. However, the suppressions of internal noise nonmonotonously change with the increasing external noise intensity. In addition, the external noise intensity for the occurrence of SREN increases with the increasing internal noise. This result is different from the performance of the external noise of NO partial pressure coupled to internal noise [47]. A simple mechanism for this difference is given.

2. Model

The stochastic model here is developed on the basis of the deterministic model [48]. Following the Langmüir–Hinshelwood mechanism, the NO + CO reaction on Pt (100) can be described by the following steps [48]:

$$\begin{aligned}
&\mathrm{CO_{gas}} \underset{k_2}{\overset{k_1}{\rightleftarrows}} \mathrm{CO_{ads}}, \quad \mathrm{NO_{gas}} \underset{k_4}{\overset{k_3}{\rightleftarrows}} \mathrm{NO_{ads}}, \quad \mathrm{NO_{ads}} \xrightarrow{k_5} \mathrm{N_{ads}} + \mathrm{O_{ads}},\\
&2\mathrm{N_{ads}} \rightarrow (\mathrm{N_2})_{\mathrm{gas}}, \quad \mathrm{CO_{ads}} + \mathrm{O_{ads}} \xrightarrow{k_6} (\mathrm{CO_2})_{\mathrm{gas}}.
\end{aligned} \tag{1}$$

The deterministic kinetics of (1) is governed by:

$$\begin{aligned}
\frac{\mathrm{d}\theta_{\mathrm{CO}}}{\mathrm{d}t} &= k_1 P_{\mathrm{CO}}(1-\theta_{\mathrm{CO}}-\theta_{\mathrm{NO}}) - k_2\theta_{\mathrm{CO}} - k_6\theta_{\mathrm{CO}}\theta_{\mathrm{O}},\\
\frac{\mathrm{d}\theta_{\mathrm{NO}}}{\mathrm{d}t} &= k_1 P_{\mathrm{NO}}(1-\theta_{\mathrm{CO}}-\theta_{\mathrm{NO}}) - k_4\theta_{\mathrm{NO}} - k_5\theta_{\mathrm{NO}}\theta_{\mathrm{empty}},\\
\frac{\mathrm{d}\theta_{\mathrm{O}}}{\mathrm{d}t} &= k_5\theta_{\mathrm{NO}}\theta_{\mathrm{empty}} - k_6\theta_{\mathrm{CO}}\theta_{\mathrm{O}},
\end{aligned} \tag{2}$$

with

$$\theta_{\mathrm{empty}} = \max\left[\left(1 - \frac{\theta_{\mathrm{CO}}+\theta_{\mathrm{NO}}}{\theta^{\mathrm{inh}}_{\mathrm{CO,NO}}} - \frac{\theta_{\mathrm{O}}}{\theta^{\mathrm{inh}}_{\mathrm{O}}}\right), 0\right],$$

where $\theta_{\mathrm{NO,CO,O}}$ stand for the absorbed coverage of NO, CO, and oxygen. These equations consist of the adsorption and desorption of NO and CO (k_1, k_2, k_3, and k_4), the dissociation of NO (k_5), and the surface reaction between adsorbed oxygen and adsorbed CO to form CO_2 (k_6). P_{CO} and P_{NO} are the respective partial pressures of CO and NO gas.

Table 1
The constants used in the model

Description	Constants	E_i (kcal/mol)	ν_i (s^{-1})
CO/NO adsorption	k_1/k_3		
CO desorption	k_2	37.5 ($\theta = 0$)	1.0×10^{14}
$CO_{ad} + O_{ad}$ reaction	k_6	14.0	2.0×10^{8}
NO desorption	k_4	37.0 ($\theta = 0$)	1.7×10^{14}
NO dissociation	k_5	28.5	2.0×10^{15}
CO/NO repulsion	k_7	24	
Inhibition coverage for NO dissociation	$\theta^{\mathrm{inh}}_{\mathrm{CO,NO}}$	0.61	
	$\theta^{\mathrm{inh}}_{\mathrm{O}}$	0.4	

The temperature dependence is expressed via Arrhenius law $k_i = \nu_i \exp(-E_i/RT)$.

Table 2
The stochastic processes and transition rates for NO reduction by CO inside one single cell on Pt(100)

Stochastic processes	Reaction rate
$N_{\mathrm{CO}} \to N_{\mathrm{CO}} + 1$,	$a_1 = k_1 P_{\mathrm{CO}}(V - N_{\mathrm{CO}} - N_{\mathrm{NO}})$,
$N_{\mathrm{CO}} \to N_{\mathrm{CO}} - 1$,	$a_2 = k_2 N_{\mathrm{CO}}$,
$N_{\mathrm{NO}} \to N_{\mathrm{NO}} + 1$,	$a_3 = k_1 P_{\mathrm{NO}}(V - N_{\mathrm{CO}} - N_{\mathrm{NO}})$,
$N_{\mathrm{NO}} \xrightarrow{\text{desorption}} N_{\mathrm{NO}} - 1$,	$a_4 = k_4 N_{\mathrm{NO}}$,
$N_{\mathrm{NO}} \xrightarrow{\text{dissociation}} N_{\mathrm{NO}} - 1$,	$a_5 = k_5 N_{\mathrm{NO}} N_{\mathrm{empty}} V^{-1}$,
$\begin{pmatrix} N_{\mathrm{N}} \to N_{\mathrm{N}} + 1, N_{\mathrm{O}} \to N_{\mathrm{O}} + 1, \\ N_{\mathrm{empty}} = \max\{[V - (N_{\mathrm{CO}} + N_{\mathrm{NO}})(0.61)^{-1} - N_{\mathrm{O}}(0.4)^{-1}]V^{-1}, 0\} \end{pmatrix}$	
$(N_{\mathrm{CO}}, N_{\mathrm{O}}) \to (N_{\mathrm{CO}} - 1, N_{\mathrm{O}} - 1)$,	$a_6 = k_6' N_{\mathrm{CO}} N_{\mathrm{O}} V^{-1}$

Note: All parameter values are listed in Table 1 with exception of replacement of k_6 by noisy rate constant k_6'.

The number of vacant sites available for NO dissociation, θ_{empty}, can be calculated from the inhibition coverage, θ_x^{inh}, for NO dissociations of each individual adsorbate. The system (2) exhibits Hopf bifurcation at $P_{\mathrm{NOh}} = 3.044 \times 10^{-7}$ mbar when $P_{\mathrm{CO}} = 3 \times 10^{-7}$ mbar. The various constants used in the equations are given in Table 1.

The adsorption energy $E_{\mathrm{ad}}^{\mathrm{CO,NO}}$ was parameterized for both gases with the same fitting parameter k_7 by

$$E_{\mathrm{ad}}^{\mathrm{CO,NO}}(\theta) = E_{\mathrm{ad}}^{\mathrm{CO,NO}}(0) - k_7\theta^2,$$

with θ denoting the sum of the CO and NO coverage, $\theta = \theta_{\mathrm{NO}} + \theta_{\mathrm{CO}}$.

For the reaction taking place on a small-size surface, since the number of reacting molecules inside the surface is very low and the random fluctuations of reactant coverage becomes considerable, the elementary steps (1) should be replaced by the elementary events and probabilities per unit time which are listed in Table 2.

The corresponding chemical Langevin equations (CLE) can be written as:

$$\begin{aligned} \mathrm{d}\theta_{\mathrm{CO}}/\mathrm{d}t &= \frac{1}{V}\left[(a_1 - a_2 - a_6) + \sqrt{a_1}\xi_1 - \sqrt{a_2}\xi_2 - \sqrt{a_6}\xi_6\right], \\ \mathrm{d}\theta_{\mathrm{NO}}/\mathrm{d}t &= \frac{1}{V}\left[(a_3 - a_4 - a_5) + \sqrt{a_3}\xi_3 - \sqrt{a_4}\xi_4 - \sqrt{a_5}\xi_5\right], \\ \mathrm{d}\theta_{\mathrm{O}}/\mathrm{d}t &= \frac{1}{V}\left[(a_5 - a_6) + \sqrt{a_5}\xi_5 - \sqrt{a_6}\xi_6\right], \end{aligned} \tag{3}$$

where V is the system size which determines the number of absorption sites on the surface; $\xi_i(t)$ $(i = 1, \ldots, 6)$ are Gaussian white noises with $\langle \xi_i(t) \rangle = 0$ and $\langle \xi_i(t)\xi_j(t') \rangle = 2Q\delta_{ij}\delta(t - t')$, with Q being noise intensity; and a_i are reaction rates associated with V and hence are related to internal noise.

To study the effect of the external noise of reaction rate constant, we consider the random variation of the rate constant k_6 for the reaction of CO and O by the modulation of white noise $\gamma(t)$:

$$k_6' = k_6[1 + D\gamma(t)], \tag{4}$$

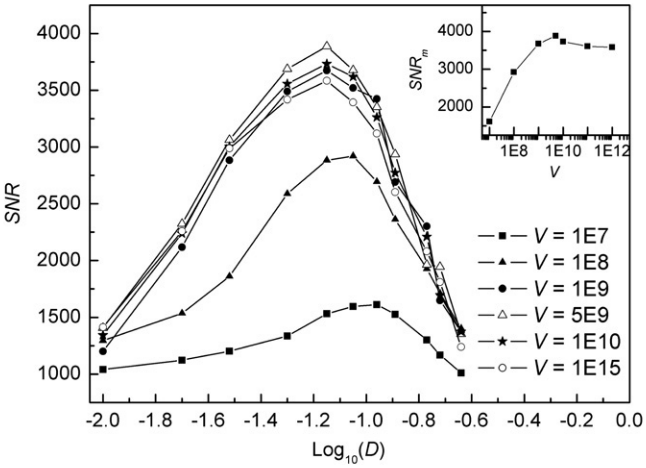

Fig. 1. Enhancement of SREN by internal noise. The peak of the SNR curve for the external noise increases with decreasing system size and reaches the maximum height at about $V = 5 \times 10^9$, and then decreases with further decreasing system size. The external noise intensity D for the SREN increases with increasing internal noise.

where $\gamma(t)$ is Gaussian white noise with $\langle\gamma(t)\rangle = 0$ and $\left\langle\gamma(t)\gamma(t')\right\rangle = 2D\delta(t-t')$, D is the noise intensity; and k_6' is the noisy rate constant. We fix $P_{\mathrm{NO}} = 3.043 \times 10^{-7}$ mbar so that the deterministic system (2) is outside but near the Hopf bifurcation point P_{NOh}. The explicit Euler algorithm with time step of 0.01 s is employed in our numerical simulations.

3. Results and discussion

The SREN and SRIN in the present model are similar to those in Ref. [47] and hence will not be studied in detail here again. We will focus on how the SREN is affected by the internal noise and the SRIN by the external noise.

3.1. Enhancement of SREN by internal noise

We first present the SREN approximately without internal noise by letting the system size be extremely large $V = 10^{15}$. The SNR curve (empty circles) for the SREN is shown in Fig. 1. As the internal noise is added and the intensity is increased, the SNR curve becomes higher (pentacles for $V = 10^{10}$), and reaches the maximal height at about $V = 5 \times 10^9$ (empty triangles). However, as the internal noise intensity is further increased, the SNR curve declines (solid circles for $V = 10^9$), falls rapidly when $V = 10^8$ (solid triangles), and becomes very low at $V = 10^7$ (solid squares), at which the internal noise intensity is considerably large. It is clearly seen that the internal noise in the range $V = 10^9$–10^{10} can enhance the SREN. However, as the internal noise is increased to $V = 10^8$, it will reduce the SREN. As it is further increased to $V = 10^7$ or less size, it will smear the external noise-induced oscillations and destroy the SREN.

It is also shown in Fig. 1 that the external noise intensity D for the SREN almost keeps unchanged when the internal noise is very small at about $\sim 10^9$ or larger. However, the value of D for the SREN increases with the increasing internal noise ($V \sim 10^8$ or less), which is opposite to the performance of the external noise of the NO partial pressure [47].

3.2. Suppression of SRIN by external noise

The SNR curve for the SRIN without external noise ($D = 0$) is presented in Fig. 2 (empty circles). As the external noise with intensity $D = 0.001$ is applied, the SNR curve falls rapidly (solid circles), showing the strong suppression of the SRIN by the external noise. As the external noise is increased, the suppression is strengthened correspondingly ($D = 0.003$ empty squares), and the SNR curve falls to the minimal height at $D = 0.005$. However, as the external noise is further increased ($D = 0.006$, 0.007), the SNR curve becomes higher (see the inset in Fig. 2). Up to $D = 0.008$, the SNR curve reaches the maximal height (empty triangles). The rise of the SNR curve implies that the suppression of the SRIN by external noise is weakened. In addition, the SNR curve for $D = 0.008$ rises again at about

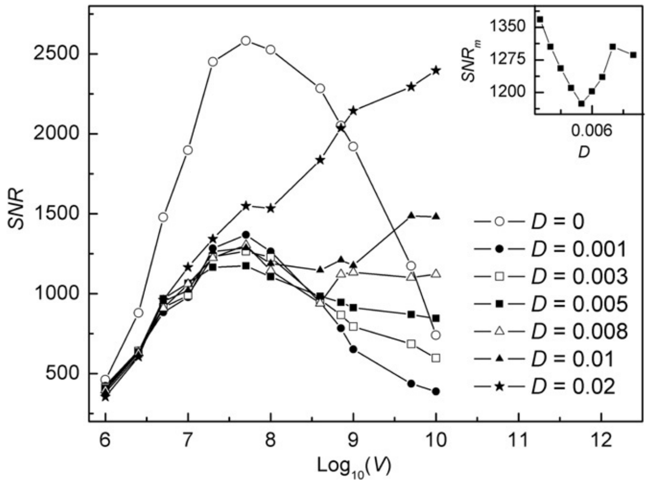

Fig. 2. Suppression of SRIN by external noise. The peak of the SNR curve for the internal noise drastically decreases as the external noise is added, meaning the SRIN is heavily suppressed by the external noise. The suppression of the SRIN changes nonmonotonously with the external noise strength, i.e., within a range of external noise strength, the higher external noise causes a lower suppression on the SRIN than the lower external noise.

$V = 5\times10^8$ and then goes to a plateau after it first arrives at the peak at about $V = 5\times10^7$ and then begins to fall, which implies that the external noise with $D = 0.008$ becomes larger compared to the small internal noise ($V > 5 \times 10^8$) so that it begins to destroy the SRIN. As the external noise is increased to $D = 0.01$ (solid triangles) and $D = 0.02$ (pentacles), the peak becomes very small and disappears, respectively, and then the SNR undergoes a plateau (not shown). This indicates the external noise becomes so large that the total noise completely destroys the SRIN.

The conclusions can be drawn from Fig. 2 that the internal noise can enhance the SREN, but the external noise of k_6 would suppress the SRIN, and the suppressions nonmonotonically vary with the changing external noise intensity, i.e., within a range of external noise intensity, the higher external noise causes a lower suppression on the SRIN than the lower external noise.

The present result for the effect of the external noise of reaction rate constant on the SRIN is much different from that in Ref. [47] in which the external noise of NO partial pressure enhances the SRIN. Although it is not yet very clear how the two kinds of external noises separately interact with the internal noise and produce such different effects, the different external noise sources and their different functions in the reaction as well as the underlying nonlinearity of the system are the main reason. It is known from Eq. (3) and Table 2 that the rate a_3 including NO partial pressure exerts positive feedback functions, but the rate a_6 involving k'_6 plays negative feedback functions in the reaction. When the SRIN occurs, the former can enhance the performance of stochastic oscillations by introducing extra dynamics which may play a crucial role as an energy source, whereas, the latter can reduce the performance of stochastic oscillations by consuming the oscillatory energy. Therefore, the external noise of NO partial pressure and of rate constant k_6 would cause different effects in the SRIN through their separate interplay with the nonlinearity of the system. This result may be of another example that different noise sources may affect the system's stochastic dynamics in different ways [49,50].

As stated in the Introduction, the SREN or SRIN, i.e., the enhancement of oscillatory signals at an optimal external or internal noise (system size), has already been observed in experiments. But the interaction of external and internal noise and its influence on the SR have not been investigated in experiment. Our findings suggest the SRIN or SREN can be enhanced or suppressed by external or internal noise, in dependence of the generation and functions of the noise term, as well as the specific nonlinearity of the system. This phenomenon can be studied in experiment by observing the change of the strength of oscillatory signal when the external noise is applied or the system size is changed. This phenomenon is interesting and is expected to be studied in future experimental work.

4. Conclusion

Based on the stochastic model of catalytic NO reduction by CO, we have studied the effects of the external noise of reaction rate constant k_6 coupled to the internal noise in the SREN or SRIN. It is shown that the internal noise can enhance the SREN, and the external noise intensity for the SREN increases with the increasing internal noise. But the external noise can suppress the SRIN, and the suppressions nonmonotonously vary with the changing external noise strength. This result is different from the effect of the external noise of NO partial pressure coupled to the internal noise. This difference may arise from the different generation mechanisms and functions of the external noise terms and the interaction of the external noise with the specific nonlinearity of the system.

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