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Internal noise-enhanced phase synchronization of coupled chemical chaotic oscillators

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Abstract

The effect of internal noise on phase synchronization of two coupled chemical chaotic oscillators is investigated numerically using chemical Langevin equations. It is found that internal noise can enhance the phase synchronization, and there exists an optimal internal noise level such that the best phase synchronization is achieved. Since the magnitude of the internal noise is changed via the variation of the system size, these phenomena also indicate the existence of an optimal system size.

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1. Introduction

Synchronization of coupled chaotic oscillators has received much attention [1, 2] over the last decade due to its fundamental importance with applications in various fields, such as laser dynamics [3], chemical and biological systems [4], electronic circuits [5] and secure communications [6]. Several kinds of synchronization have been found: complete synchronization [7], generalized synchronization [8], lag synchronization [9] and phase synchronization (PS) [10]. PS can be achieved with a smaller coupling strength than that for complete synchronization, and when the two non-identical systems reached the phase synchronization, their amplitudes can remain chaotic and uncorrelated with each other [1].

Very recently, the roles of noise in the synchronization of chaotic systems have attracted growing attention. The main reason is that, being well accepted now, noise can play rather counterintuitive, constructive roles in affecting the dynamic behaviours of nonlinear systems. The most famous phenomenon is stochastic resonance (SR), which shows that there exists a 'resonant' noise intensity at which the response of the system to a periodic force is maximally ordered. Similarly, constructive roles of noise in the synchronization of nonlinear oscillators have also been found [11].

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However, most of these studies so far have only accounted for *ad hoc external* noise. Usually the chaotic dynamics of the system is described by deterministic differential equations, and noise terms are added to the equations directly, multiplicatively or additively. The properties of the external noise, such as intensity or correlation time, are assumed to be controllable parameters and have no relevance to the system's dynamic features or the system's size. But for chemical reactions in small-scale systems, such as those biochemical reactions taking place in living cells [12] or catalytic reactions happening on nano-scale crystal surfaces [13], another source of noise, the *internal* noise, must be taken into account. Such internal noise is caused by the random birth and death events of chemical reacting species, which is an intrinsic feature of any chemical reaction. Unlike external noise, the internal noise is not controllable, and it depends on the details of the reaction dynamics as well as the system size. It is generally accepted that the strength of the internal noise scales as $1/\sqrt{V}$, where V is the system size. In the macroscopic limit when V is infinite, the internal noise can be ignored and one recovers the deterministic description of the system's dynamics. But for systems of small sizes, the number of the reacting molecules could be low and the internal noise becomes crucial. Therefore, an interesting question, i.e., how the internal noise affects the system's dynamics, naturally arises.

In the present work, we have investigated how the internal noise would influence the synchronization of two coupled chemical chaotic oscillators, namely, the chemical Rössler oscillators. One should note that there are a lot of real biochemical reaction systems as well as catalytic reaction systems that show chaotic behaviours [14]. Synchronization of such real chaotic systems might be of great importance, for instance, experimental evidence suggests that the synchronization of large populations of neurons shapes the processing of information by the brain [15]. Although it should be more useful to study such 'real' chaotic models, we have chosen the Rössler model here due to its simplicity for the construction of stochastic models, and the fact that the effect of external noises on the original Rössler model has been well studied [16]. We believe that the study of such a simplified model could also provide some general results, which makes the first step towards further investigations of real complex systems.

2. The chemical Rössler model

The well-known Rössler model contains three ordinary differential equations determining the time evolution of three state variables. Such a model is constructed mathematically, and it is not related to any chemical reactions. Therefore, this 'original' Rössler model is not suitable to study the effects of internal noise, although it has been widely used to study the roles of external noise or disorder on synchronization. The main issue is that a mathematical state variable can be negative, while the state variables for a chemical system, such as the concentrations, can never be less than zero. However, by using a nonlinear transformation [17] the original Rössler equations can be translated into a new form that allow only nonnegative state variables and preserve the dynamical features of the original Rössler system. Furthermore, the new equations can be interpreted on the base of mass action law as some elementary chemical reactions. Hence, one obtains a chemical Rössler system, where internal noises are inherent in the stochastic elementary reaction steps. There are, in total, nine steps as follows:

(1) $A_1 + X + 2Z \xrightarrow{k_1} X + 3Z$	$(2) X + Y \xrightarrow{k_2} 2Y$	$(3) A_2 + X + Z \xrightarrow{k_3} Z + P_1$
$(4) A_3 + X + Z \xrightarrow{k_4} X + P_2$	(5) $A_4 + 2Y \xrightarrow{k_5} 3Y$	(6) $2Z \xrightarrow{k_6} P_3$
(7) $A_5 + X \xrightarrow{k_7} 2X$	(8) $Y \xrightarrow{k_8} P_4$	$(9) A_6 + Z \xrightarrow{k_9} 2Z$



Figure 1. Chemical Rössler attractor (projected onto *x*–*y* plane). Parameters: $k_1 = 1$, $k_2 = 1$, $k_3 = 1$, $k_4 = 20$, $k_5 = 0.2$, $k_6 = 12.85$, $k_7 = 40$, $k_8 = 24$, $k_9 = 514.2$.

where parameters k_i (i = 1, 2, ..., 9) are reaction rate constants. Concentrations of species A_i (i = 1, 2, ..., 6) and P_i (i = 1, 2, ..., 4) are assumed to be constants in order to make the system far out of equilibrium. If the reactor size is infinite and it is a well-stirred ideal mixture, then internal noise can be ignored and the dynamical behaviours can be described by a set of deterministic differential equations as follows:

$$dx/dt = k_7 x - k_2 x y - k_3 x z$$

$$dy/dt = k_2 x y + k_5 y^2 - k_8 y$$

$$dz/dt = k_9 z - 2k_6 z^2 - k_4 x z + k_1 x z^2.$$
(1)

In the above equations, x, y and z are concentrations of species X, Y and Z, respectively, concentrations of A_i (i = 1, 2, ..., 6) have been incorporated into the reaction rate constants k_1 , k_3 , k_4 , k_5 , k_7 and k_9 . We integrated equations (1) using the fourth-order Runge–Kutta algorithm with a fixed step size of h = 0.0001. The system's behaviour is shown in figure 1, which is qualitatively similar to the original Rössler system, but the centre of the chaotic attractor now moves from (0, 0) to (20, 20).

To account for the effect of internal noise on phase synchronization, we coupled two non-identical chemical Rössler systems by linear symmetric diffusion of x. Each system is of finite size, hence the deterministic description is not valid. In [18], Gillespie demonstrated that the stochastic dynamics of a chemical reaction system may be well approximated by a set of chemical Langevin equations (CLEs), which show how the internal noises are related to the system size V. Specifically, the CLEs for two coupled chemical Rössler systems are

$$dx_{1}/dt = (k_{7}x_{1} - k_{2}x_{1}y_{1} - k_{3}x_{1}z_{1}) + 1/\sqrt{V} [\sqrt{k_{7}x_{1}}\xi_{7}(t) - \sqrt{k_{2}x_{1}y_{1}}\xi_{2}(t) - \sqrt{k_{3}x_{1}z_{1}}\xi_{3}(t)] + \varepsilon(x_{2} - x_{1}) dy_{1}/dt = (k_{2}x_{1}y_{1} + k_{5}y_{1}^{2} - k_{8}y_{1}) + 1/\sqrt{V} [\sqrt{k_{2}x_{1}y_{1}}\xi_{2}(t) + \sqrt{k_{5}y_{1}^{2}}\xi_{5}(t) - \sqrt{k_{8}y_{1}}\xi_{8}(t)]$$

 $dz_{1}/dt = (k_{9}z_{1} - 2k_{6}z_{1}^{2} - k_{4}x_{1}z_{1} + k_{1}x_{1}z_{1}^{2})$ $+ 1/\sqrt{V} [\sqrt{k_{9}z_{1}}\xi_{9}(t) - \sqrt{2k_{6}z_{1}^{2}}\xi_{6}(t) - \sqrt{k_{4}x_{1}z_{1}}\xi_{4}(t) + \sqrt{k_{1}x_{1}z_{1}^{2}}\xi_{1}(t)]$ $dx_{2}/dt = (k_{16}x_{2} - k_{11}x_{2}y_{2} - k_{12}x_{2}z_{2})$ $+ 1/\sqrt{V} [\sqrt{k_{16}x_{2}}\xi_{16}(t) - \sqrt{k_{11}x_{2}y_{2}}\xi_{11}(t) - \sqrt{k_{12}x_{2}z_{2}}\xi_{12}(t)] + \varepsilon(x_{1} - x_{2})$ $dy_{2}/dt = (k_{11}x_{2}y_{2} + k_{14}y_{2}^{2} - k_{17}y_{2})$ $+ 1/\sqrt{V} [\sqrt{k_{11}x_{2}y_{2}}\xi_{11}(t) + \sqrt{k_{14}y_{2}^{2}}\xi_{14}(t) - \sqrt{k_{17}y_{2}}\xi_{17}(t)]$ $dz_{2}/dt = (k_{18}z_{2} - 2k_{15}z_{2}^{2} - k_{13}x_{2}z_{2} + k_{10}x_{2}z_{2}^{2})$ $+ 1/\sqrt{V} [\sqrt{k_{18}z_{2}}\xi_{18}(t) - \sqrt{2k_{15}z_{2}^{2}}\xi_{15}(t) - \sqrt{k_{13}x_{2}z_{2}}\xi_{13}(t) + \sqrt{k_{10}x_{2}z_{2}^{2}}\xi_{10}(t)].$ (2)

Here, the subscripts 1 and 2 of the state variables (x, y, z) represent the systems 1 and 2, respectively. The parameters are $k_1 = 1$, $k_2 = 1.02$, $k_3 = 1$, $k_4 = 20$, $k_5 = 0.2$, $k_6 = 12.85$, $k_7 = 40.4$, $k_8 = 24.4$, $k_9 = 514.2$ for the first system and $k_{10} = 1$, $k_{11} = 0.98$, $k_{12} = 1$, $k_{13} = 20$, $k_{14} = 0.2$, $k_{15} = 12.85$, $k_{16} = 39.6$, $k_{17} = 23.6$, $k_{18} = 514.2$ for the second one; $\xi_i = 1, \ldots, 18(t)$ are Gaussian white noises with $\langle \xi_i(t) \rangle = 0$ and $\langle \xi_i(t) \xi_j(t') \rangle = \delta_{ij} \delta(t - t')$, and ε is the coupling strength. The above equations are the same as the deterministic equations (1) if the second terms in the brackets at the right side are absent. Therefore, these terms actually result from the internal noises. It is clear that the magnitude of the internal noises is proportional to $1/\sqrt{V}$, and they depend not only on the concentrations of the molecular species but also the reaction rate constants. The internal noise is not solely additive, but essentially additive, depending on the state of the system.

From the CLEs we know the magnitude of the internal noises can be changed via the change of V or the variation of a certain control parameters as shown in equations (2). But the change of a certain control parameter also changes the deterministic kinetics. However, to address the influence of internal noise on the system's dynamics, we need to scan the magnitude of the internal noise over a relatively wide range, and keep the corresponding deterministic kinetics unchanged. Therefore in the present work, the magnitude of the internal noise will be changed via the change of V.

3. Results and discussion

To measure the phase synchronization, we need to introduce amplitude and phase variables to the system. There are several kinds of definitions of 'phase' in the literature [19], such as the natural phase, linear interpolating phase, Hilbert phase, discrete phase and so on. Form figure 1, we know that the centre of the chemical Rössler attractor is (20, 20), hence we can define the amplitude and phase as $A_i^2 = (x_i - 20)^2 + (y_i - 20)^2$ and $\tan \phi_i = \frac{y_i - 20}{x_i - 20}$ [20]. One notes that the coupling between the two Rösslers does not greatly change the position of the centre, such that the phase definition remains valid for the coupled system. Furthermore, we define the average frequency difference $\Delta \Omega = |\langle \Delta \dot{\phi} = \dot{\phi}_1 - \dot{\phi}_2 \rangle|$, which will be used to quantitatively measure the synchronization of the two systems. Here, $\langle \cdot \rangle$ stands for the averaging over time and $|\cdot|$ denotes the absolute value. Note that a smaller $\Delta \Omega$ means a better phase synchronization.

To begin, we perform numerical calculations of equations (2) in the deterministic case $V \rightarrow \infty$ when the internal noise can be ignored. The average frequency $\Delta\Omega$ versus ε



Figure 2. The average frequency difference $\Delta\Omega$ versus ε for $V \to \infty$ is plotted. There exists a transition point $\varepsilon_{ps} \approx 0.74$. The total time we calculated is 10^8 time steps.



Figure 3. The curves of the average frequency difference $\Delta\Omega$ versus $1/\sqrt{V}$ for $\varepsilon = 0.65, 0.71$ and 0.77, respectively. The total time we calculated is 10^8 time steps.

is displayed in figure 2, where it shows that $\Delta\Omega$ decreases along with the increase of the coupling strength. At a transition point $\varepsilon_{ps} \approx 0.74$, $\Delta\Omega$ decreases to nearly zero that refers to a complete phase synchronization. In the present work, we will focus on the effect of internal noise when ε is tuned close to the transition point.

Figure 3 illustrates the dependence of $\Delta\Omega$ on $1/\sqrt{V}$ for three difference coupling strengths $\varepsilon = 0.65$, 0.71 and 0.77, taking into account the internal noise. One notes that $1/\sqrt{V}$ here qualitatively measures the relative magnitude of the internal noise. For $\varepsilon = 0.65$ and 0.71, there exists an optimal internal noise strength, at which $\Delta\Omega$ has a minimum. But for $\varepsilon = 0.77$, internal noise is always disadvantageous to the phase synchronization. The curves also exhibit a smaller $\Delta\Omega$ for a larger ε when $1/\sqrt{V}$ is given. We have scanned the magnitude of internal noise and the coupling strength over a relatively wide range, and similar results are obtained as shown in figure 4.

The existence of an optimal internal noise level for phase synchronization can also be reflected in the temporal behaviour of $\Delta \phi = \phi_1 - \phi_2$. Figure 5 shows such an example for



Figure 4. The average frequency difference $\Delta\Omega$ as a function of ε and $1/\sqrt{V}$. The total time we calculated is 10^8 time steps.



Figure 5. The evolution of phase difference for $\varepsilon = 0.071$ with $1/\sqrt{V} = 0.0005$, 0.0009 and 0.0013, respectively. The unit of *t* is 10^4 time steps.

 $\varepsilon = 0.71$. The curves in figure 5 consist of many synchronization epochs connected by sharp phase slips. On the epochs, $\Delta\phi$ remains constant such that $\Delta\dot{\phi} = 0$, contributing nothing to $\Delta\Omega$. Therefore, $\Delta\Omega$ mainly comes from the sharp change of $\Delta\phi$ during the phase slips. For relatively small and large internal noise strengths, e.g., $1/\sqrt{V} = 0.0005$ or 0.0013, there are many phase slips between which are short epochs of phase synchronization. While at an optimal internal noise level, e.g., $1/\sqrt{V} = 0.0009$, the synchronization epochs are longer and the phase slips occur much less frequently.

As being stated in [16], the interplay between internal noise and unstable periodic orbits (UPOs) is important for the understanding of internal noise-enhanced PS. As we know, there are many UPOs embedded in the chaotic attractors and chaotic trajectories can stay close to a certain UPO for some time. PS of coupled chaotic systems can be viewed as phase-locking of a number of UPOs, and phase slips are indeed generated by unlocked UPOs [16]. In the

complete phase synchronization region ($\varepsilon > \varepsilon_{ps}$), all pairs of UPOs are mutually locked to each other. When the coupling strength ε decreases across the transition point ε_{ps} , some pairs of UPOs will loss phase-locking. Therefore, if the coupling strength is close to and less than the transition point, internal noise is expected to play interesting roles.

If the two systems are phase-locked on a pair of UPOs, of which the lifetimes are long enough, then phase synchronization could occur and the phase difference $\Delta\phi$ would remain on the epoch (see figure 5). If each system can follow a UPO for enough 'long' time and the two UPOs are not locked to each other, then $\Delta\phi$ may change abruptly and phase slip occurs. Consequently, noise has two effects [16]: (i) it prevents the two systems to stay close to unlocked UPOs for long enough time to allow a phase slip to occur; (ii) it generates fluctuation in the return times (return time: the time between two successive returns of the chaotic trajectory to a Poincaré section) and may induce phase slip of locked orbits. The degree of PS is enhanced when (i) is dominant over (ii) at weak noise level, while it is degraded when (ii) becomes dominant at larger noises. Thus, there is an optimal internal noise intensity when ε is smaller than ε_{ps} because of the competition of these two effects. While in the phase synchronization region where $\varepsilon > \varepsilon_{ps}$, internal noise only has the second effect is disadvantageous to PS.

4. Conclusion

In conclusion, we have studied the influence of internal noise on phase synchronization of two chemical Rössler chaotic oscillators by numerical simulations of chemical Langevin equations. It is found that a proper internal noise is advantageous to phase synchronization. Since the magnitude of internal noise is changed via the variation of the system size, these phenomena also indicate the existence of an optimal system size.

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