Partial synchronization of relaxation oscillators with repulsive coupling in autocatalytic integrate-and-fire model and electrochemical experiments

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Experiments and supporting theoretical analysis is presented to describe the synchronization patterns that can be observed with a population of globally coupled electrochemical oscillators close to a homoclinic, saddle-loop bifurcation, where the coupling is repulsive in the electrode potential. While attractive coupling generates phase clusters and desynchronized states, repulsive coupling results in synchronized oscillations. The experiments are interpreted with a phenomenological model that captures the waveform of the oscillations (exponential increase) followed by a refractory period. The globally coupled autocatalytic integrate-and-fire model predicts the development of partially synchronized states that occur through attracting heteroclinic cycles between out-of-phase two-cluster states. Similar behavior can be expected in many other systems where the oscillations occur close to a saddle-loop bifurcation, e.g., with Morris-Lecar neurons.

PACS numbers: 05.45.Xt, 82.40.Bj

Many oscillatory processes underlie the functioning of important biological and engineered systems. The waveform of the oscillation of a variable (e.g., concentration of substances) has strong impact on the overall behavior, e.g. on how the oscillations synchronize together. The waveform can be smooth, nearly sinusoidal, or relaxation type, where slow variations are followed by a quick spike. In this paper, we performed experiments with a chemical oscillatory system, where the waveform had strong relaxation character, and show that such a system, in contrast with the previously studied smooth oscillation, can produce synchronization with repulsive coupling among the variables. The experiments are interpreted with a simple mathematical model, where the relaxation character of the waveform can be tuned to generate complex synchronization patterns.

I. INTRODUCTION

The widespread occurrence of different types of synchronization patterns of nonlinear dynamical systems calls for theoretical description using simplified, generic models [1–3]. The development of such models depends on local nonlinear features of the oscillating units and the type of interactions. When the interactions are global and weak, the oscillations can often be described with phase models,

$$\frac{d\phi_i}{dt} = \omega_i + \sum_{j=1}^{N} \Gamma(\phi_i - \phi_j),$$

where $\phi_i (i = 1 \ldots N)$ and $\omega_i$ are the phase and the natural frequency of the $i$-th oscillator, respectively, and $N$ is the number of oscillators [2]. The function $\Gamma$ is referred to as the phase interaction function, obtained as the convolution of the phase sensitivity function $Z(\phi)$ and the waveform of an interacting agent. The function $Z(\phi)$ is proportional to the phase response curve, which has been measured in many systems including chemical and biological oscillators. The central component in such a phase model is the functional form of $\Gamma(\phi)$. For example, close to a Hopf bifurcation with coupling that occurs through an additive term of variable differences, $\Gamma$ is a sinusoidal function, possibly shifted with a constant term, as in the Kuramoto-Sakaguchi model [2]. This property results from the fact that both $Z$ and the waveform are sinusoidal [2]. Further away from the Hopf bifurcation, higher harmonics can occur in $Z$, as theoretically shown [4] and observed in many chemical and biological oscillators [5–10]. Correspondingly, higher harmonics appears also in $H$ giving rise to phase cluster dynamics [4]. Because of the relative simplicity of the mathematical structure of phase models, very often analytical solutions exist for synchronization patterns.

Many oscillations in nature, for example, in chemistry and neurophysiology, have more complex shapes. The slow, exponential decaying waveform, corresponding to the charging of the membrane potential in biology, motivated the development of integrate-and-fire (IF) types of models. In IF models after the process is complete, there is quick, often instantaneous discharge that allows the process to restart. Such models, which typically describe the behavior close to bifurcation (e.g. homoclinic saddle loop (SL) bifurcation or saddle-node bifurcation of infinity period (SNIPE)) [11], can generate rich dynamics in networks, that include synchronization [12], asynchronous dynamics [13], clustering [14], or chimera states [15]. Systems close to a SNIPE or SL bifurcation, can generate a refractory period: the discharging

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process is not instantaneous, but occurs over a relatively short interval during which the system is insensitive to external perturbations [16].

In this paper, we design an integrate-and-fire type of model for the description of synchronization patterns of an electrochemical oscillator close to SL bifurcation. The experiment, performed with a repulsive coupling of a population of electrochemical oscillators, exhibits a synchronized state. The extent of the synchronization is investigated as a function of distance from the SL bifurcation. The experiments are interpreted with an autocataytic integrate-and-fire (AIF) model, adjusted for the exponentially increasing waveform for the experiments. The AIF model is analyzed using a phase model description. The model analysis reveals the type of synchronized oscillations, clusters, and partially synchronized states.

II. EXPERIMENTAL RESULTS AND ANALYSIS

We carried out experiments to explore the synchronization behavior with $N = 64$ electrochemical relaxation oscillators close to a homoclinic bifurcation. The system consists of 64 metal wires; the rate of metal dissolution (currents of the electrodes) at constant circuit potential ($U$) is measured. The currents of the electrodes become oscillatory through a supercritical Hopf bifurcation point at $U = 1.0$ V; the oscillations are smooth near the Hopf bifurcation point. As the potential is increased further, relaxation oscillations are seen that disappear into a steady state through a homoclinic bifurcation at about $U = 1.31$ V [9]. In experiments with the circuit potential potentials close to the homoclinic bifurcation point, some electrodes passivated during the experiments; in this case these electrodes were disconnected and the experiments were continued with a smaller number of oscillators as indicated in the figures. In all reported experiments $N \geq 47$. The electrodes are coupled with a combination of resistors coupled in series ($R_s$) and in parallel ($R_p$); the imposed coupling strength is $K = NR_s/R_p$ [9]. Negative coupling was induced with the application of negative series resistance (built in a PAR 273A potentiostat) [4].

In a previous publication [4], we showed that close to the Hopf bifurcation with strong negative coupling the weak nonlinearities can generate cluster states. Further away from the Hopf bifurcation the system showed desynchronized behavior.

Here we focus on behavior at even larger circuit potentials, where relaxation oscillations occur that cease through a homoclinic bifurcation. Figure 1 shows that under these conditions the system exhibits nearly synchronized behavior. While the oscillators do not spike completely together (see Fig. 1a and c for time series in current vs time and grayscale plots, respectively), they form a tightly synchronized cluster. This synchronization can be also seen in the phase snapshot in Fig. 1b, where the phases of the oscillators, $\phi_j(t)$, were calculated with the Hilbert transform approach [3, 17]. The extent of synchronization can be quantitatively characterized using the average Kuramoto order parameter $R(t) = \frac{1}{N} \sum_{j=1}^{N} e^{i \phi_j(t)}$ after a transient. We have $R(t) = 1$ when all the phases take the same value (in-phase synchrony) and $R(t) = 0$ for a uniform phase distribution including the balanced cluster states. As shown in Fig. 1d, the synchronized oscillations generate large order parameter close to 1.

We carried out a series of experiments in which the circuit potential was increased and the averaged Kuramoto order was calculated (see Fig. 2). As the circuit potential (and thus the relaxation character) of the oscillators was increased, an increase in the Kuramoto order parameter was observed. Interestingly, the increase of the order is not very sharp, as, for example, could be expected from
a bifurcation that leads to a stable one-cluster state. In the inset, it is shown that at $U = 1.25$ V, in the state space an enhanced synchronization is present.

A. Phase model analysis with experimentally obtained phase interaction function

For better experimental characterization of the synchronization transition with increasing the circuit potential, we performed a phase model analysis, where the phase interaction function was constructed from experimentally measured phase response functions [9, 18]. Using the phase model, the stability of the different cluster states can be calculated for a population of globally coupled oscillators. For a large interval in the moderately relaxational oscillation region, $(1.25 \, \text{V} < U < 1.28 \, \text{V})$, the experimentally measured interaction function predicted desynchronization, however, a one cluster state with elevated value of order parameter was experimentally observed (see Fig. 2).

The difference between a population generating low and elevated $\langle R \rangle$ can be demonstrated with experimentally determined coupling functions for $U = 1.225$ V and $U = 1.265$ V [9, 18]. The measured $\Gamma(\phi)$ functions were expressed as a Fourier series up to tenth harmonics, which are shown in Fig. 3(a). By directly simulating the phase model with these $\Gamma(\phi)$ functions, we obtained dynamics similar to experimental ones, as shown in Fig. 3(b) and (c). Further using the $\Gamma(\phi)$ functions, we checked the linear stability (see Appendix A) of the balanced $n$-cluster states ($n = 1, \ldots, 10$) [19], indicating that the states with $n = 7, 8, 9$ are stable for $U = 1.225$ V whereas no state is stable for $U = 1.265$ V. The former result explains the almost uniform distribution of phases observed for $U = 1.225$ V. However, the latter can not account for the emergence of a type of one cluster state observed for $U = 1.265$ V. In order to explore the emergent collective synchronization approaching the homoclinic bifurcation, we develop a phenomenological model in the next section.

III. AUTOCATALYTIC INTEGRATE-AND-FIRE MODEL

A. Model

We construct a simple, one-dimensional model for oscillations close to a homoclinic bifurcation. This model is motivated by the waveform of many chemical and biological oscillators composed of an ‘excitatory’ phase with exponential increase followed by a usually sharp decrease with a refractory period, as observed in Fig. 1a. Therefore, a coupled oscillator is described by

$$\frac{dv}{dt} = v + K p(t).$$

![FIG. 3: Numerical results using the phase model with experimentally measured coupling function. (a) phase interaction function measured experimentally for $U = 1.225$ V and $U = 1.265$ V. (b) Time-series of the order parameter $R(t)$. (c,d) Snapshots of the phases at $t = 40$ for $U = 1.225$ V and $U = 1.265$ V, respectively. The same random initial condition was employed both for $U = 1.225$ V and $U = 1.265$ V.](#)

where $v = v(t)$ is the state variable, $K$ is the coupling strength, and $p(t)$ is an external input describing the influence from other oscillators. Note that because the waveform is generated by a positive feedback mechanism in Eq. (2), we call the integrate-and-fire model ‘autocatalytic’.

When $v$ reaches 1, its value is smoothly reset to parameter $a$ ($0 < a < 1$) by obeying

$$v = e^{-b(t - t_{\text{fire}})},$$

where $t_{\text{fire}}$ is the latest time at which $v$ becomes 1. Here, we have assumed that the oscillator is not influenced by other oscillators (or external forces) during the resetting process, i.e., in an absolute refractory period. A typical time series in the absence of coupling (i.e., $K = 0$) is shown in Fig. 4(a). In this model, parameters $a$ and $b$ characterize the intrinsic dynamical property of an oscillator. It is more convenient to characterize the relaxation character of the oscillator by using the excitatory period $\tau_e$ and refractory period $\tau_r$, given by

$$\tau_e = -\ln a,$$

$$\tau_r = -\frac{\ln a}{b}.$$  

For convenience, we denote the intrinsic period by

$$T = \tau_e + \tau_r.$$  

Larger $\tau_e$ values compared to $\tau_r$ indicate stronger relaxation character and closer distance to the homoclinic bifurcation (at which $\tau_e$ becomes infinite).
B. Phase Reduction

As a coupled oscillator system, we consider
\[ \frac{dv}{dt} = v + K(v' - v), \]  
(7)

where \( v(t) \) describes the state of an interacting oscillator. The coupling term \( K(v' - v) \) describes diffusive coupling. In the context of electrochemical and neural dynamics, \( v \) and \( K \) describes the electric potential and the conductance, respectively. Although we have included only one interacting oscillator described by \( v' \) in Eq. (7) for simplicity, we can consider a network of interacting oscillators by replacing \( v' - v \) with \( \sum_j (v_j - v) \), where \( v_j \) are the state variables of interacting oscillators.

The phase model corresponding to this model can be calculated analytically. We first define the phase \( \phi(v(t)) \) as a function of the state \( v(t) \) such that \( \frac{d\phi}{dv}(v(t)) = 1 \) for \( K = 0 \), i.e.
\[ \phi(v) = \begin{cases} 
\ln v + \tau_e & (0 \leq \phi < \tau_e, \text{excitatory phase}), \\
-\ln v + \tau_e & (\tau_e \leq \phi < T, \text{refractory phase}). 
\end{cases} \]  
(8)

By solving reversely, we obtain
\[ v(t) = \tilde{v}(\phi(t)), \]  
(9)

where
\[ \tilde{v}(\phi) = \begin{cases} 
e^{-\frac{\phi - \tau_t}{\tau_e}} & (0 \leq \phi < \tau_e), \\
e^{\frac{\phi - \tau_t}{\tau_e}} & (\tau_e \leq \phi < T). 
\end{cases} \]  
(10)

We now derive the dynamical equation of \( \phi(t) \). For the excitatory phase, using \( \frac{d\phi}{dv} = \frac{d\phi}{dv} \frac{dv}{dt} \) with Eqs. (2) and (9), we obtain
\[ \frac{d\phi}{dt} = 1 + K\tilde{Z}(\phi) \{ \tilde{v}(\phi') - \tilde{v}(\phi) \}, \]  
(11)

where \( \tilde{Z}(\phi) = e^{\tau_e - \phi} \) and \( \phi'(t) \) is the phase of the state \( v'(t) \). For the refractory phase, we have \( \dot{\phi} = 1 \). Altogether, we obtain Eq. (11) with \( \tilde{Z} \) redefined as
\[ \tilde{Z}(\phi) = \begin{cases} 
 e^{\tau_e - \phi} & (0 \leq \phi < \tau_e), \\
0 & (\tau_e \leq \phi < T). 
\end{cases} \]  
(12)

The function \( \tilde{Z}(\phi) \) is called the phase sensitivity, which describes the strength and sign of response of the phase to a perturbation applied to the oscillator. This function is defined as \( \tilde{Z}(\phi) = \left( \frac{dv}{d\phi} \right)^{-1} \), i.e. \( \left( \frac{dv}{d\phi} \right)^{-1} \) in the unperturbed system. This implies that if the perturbation is given to the oscillator when it evolves slowly, i.e., when \( v \) is small in the AIF model, the phase response is large. Therefore, \( \tilde{Z}(\phi) \) decreases with increasing \( \phi \), as described in Eq. (12).

For \( K \ll 1 \), we may further reduce Eq. (11) to a more tractable equation, given as
\[ \dot{\phi} = 1 + K\tilde{\Gamma}(\phi - \phi'), \]  
(13)

where the coupling function \( \tilde{\Gamma} \) is obtained by by averaging the right hand side of Eq. (11) over the period \( T \) [2], i.e.,
\[ \tilde{\Gamma}(\phi) = \frac{1}{T} \int_0^T \tilde{Z}(\phi + \theta) \{ \tilde{v}(\theta) - \tilde{v}(\phi + \theta) \} \, d\theta. \]  
(14)

For \( \tau_e > \tau_t \), which we assume henceforth, we obtain
\[ \tilde{\Gamma}(\phi) = \begin{cases} 
\tilde{\Gamma}_1(\phi) - \tau_e)/T & (0 \leq \phi < \tau_e, \\
\tilde{\Gamma}_2(\phi) - \tau_e)/T & (\tau_t \leq \phi < \tau_e, \\
\tilde{\Gamma}_3(\phi) - \tau_e)/T & (\tau_e \leq \phi < T), \end{cases} \]  
(15)

where
\[ \tilde{\Gamma}_1(\phi) = \frac{\tau_e}{T} e^{\frac{\phi}{T}} + \left( -\phi + \tau_e - \frac{\tau_e}{T} \right) e^{-\phi}. \]  
(16)
\[ \tilde{\Gamma}_2(\phi) = \frac{\phi - \tau_t + \frac{\tau_e}{T}}{T} e^{\tau_e - \phi} + \left( -\phi + \tau_e - \frac{\tau_t}{T} \right) e^{-\phi}, \]  
(17)
\[ \tilde{\Gamma}_3(\phi) = \frac{\phi - \tau_t + \frac{\tau_e}{T}}{T} e^{\tau_e - \phi} - \frac{\tau_t}{T} e^{-\frac{\phi}{T}(T-\phi)}. \]  
(18)

Alternatively, by introducing the phase \( \phi (0 \leq \phi < 2\pi) \) as
\[ \phi = \omega \varphi, \]  
(19)

where \( \omega = \frac{2\pi}{T} \), we obtain
\[ \dot{\phi} = \omega + K\Gamma(\phi - \phi'), \]  
(20)

where
\[ \Gamma(\phi) = \omega \tilde{\Gamma}\left( \frac{\phi}{\omega} \right). \]  
(21)

For both Eqs. (13) and (20), \( |K| \) values only determine the time scale, thus we may set \( K = 1 \) or \( K = -1 \) for positive and negative coupling, respectively, without loss of generality. In Fig. 5, we show typical \( Z(\phi) \) and \( \Gamma(\phi) \) functions. There is a notable similarity between \( \Gamma(\phi) \) functions obtained experimentally and theoretically (Figs. 3(a) and 5(c)). In particular, there is a rapid growth in a region of small \( \phi \), and the slope is steeper for a more relaxation oscillator, i.e., higher \( U \) and \( \tau_e \) values.
C. Analysis

Now we analyze a system of globally coupled oscillators. The system is given as

\[
\dot{v}_i = v_i + \frac{K}{N} \sum_{j=1}^{N} (v_j - v_i),
\]

and its corresponding phase model is obtained as

\[
\dot{\phi}_i = \omega + \frac{K}{N} \sum_{j=1}^{N} \Gamma(\phi_i - \phi_j),
\]

where \(v_i\) and \(\phi_i\) (\(i = 1, \ldots, N\)) is the state and phase of oscillator \(i\), respectively. As described in Appendix A, the local stability of the balanced \(n\)-cluster states is determined by the nontrivial maximum eigenvalue [19]. Figure 6 shows a stability diagram. Here, we only consider \(n \leq 10\) for simplicity. With positive coupling (\(K > 0\)), at low values of \(\tau_c\) only the 1-cluster state is stable. As the relaxation character increases 2, 3, 4-cluster states become progressively stable. With negative coupling (\(K < 0\)), stable cluster states exist for small \(\tau_c\) values. However, all cluster states are predicted to be unstable for large \(\tau_c\) values. Numerical simulations with the AIF model indicate that the average Kuramoto order parameter \(\langle R \rangle\) is vanishingly small for small \(\tau_c\) values.

There, in accordance with the stability analysis, balanced cluster states were observed. However, \(\langle R \rangle\) begins to increase at \(\tau_c \approx 2.0\) and takes a large value (close to unity) as \(\tau_c\) increases.

To understand the emergence of synchrony, we first focus on its onset at \(\tau_c \approx 2.0\). As shown in Fig. 6(b), only the balanced 7-cluster state is stable just below \(\tau_c = 2.0\) and the state loses stability at \(\tau_c \approx 2.0\). As also indicated in Fig. 6(b), the maximum nontrivial eigenvalue is imaginary and its mode is associated with inter-cluster fluctuations (see Appendix A). This implies that the balanced 7-cluster state loses its stability through a Hopf bifurcation and the distribution of relative phases \(\phi_i - \phi_1\) starts to oscillate after that. Figure 8 shows the time series of relative phases for different \(\tau_c\) values. At \(\tau_c = 1.8\) [Fig. 8(a)], the system converged to the balanced 7-cluster state from a random initial condition, as predicted. At \(\tau_c = 2.0\) [Fig. 8(b)], where no balanced cluster states are predicted to be stable, the system converged to a slightly scattered balanced cluster state, in which relative phases between clusters oscillate with time. This state can be interpreted as a similar state to those that bifurcate from the balanced \(n\)-cluster states via Hopf bifurcations.

For larger \(\tau_c\) values, no well-defined clusters are observed. Instead, the oscillators form a noisy cloud similar to Fig. 3(d) in spite of the instability of the one-cluster state. Figure 8(c) shows a typical time series of relative phases, where the center of the cloud travels with time like a wave; i.e., each oscillator enters and exits from a cloud repeatedly. When \(\tau_c\) is further increased, \(\langle R \rangle\) suddenly jumps at \(\tau_c \approx 2.8\), as shown in Fig. 7. Figure 8(d) shows typical time series of relative phases for \(\tau_c > 2.8\). The oscillators split into two groups, and each group repeats aggregation and breakup.

\[\langle R \rangle \approx \sqrt{\frac{1 + \cos \Delta \phi}{2}},\]

where \(\Delta \phi\) is the phase difference between two clusters. \(\Delta \phi\) is obtained as the solution to \(\Gamma(\Delta \phi) = \Gamma(-\Delta \phi)\) (see Appendix B), which is typically small, e.g., \(\Delta \phi \approx 0.3\) rad at \(\tau_c = 0.3\), thus high \(\langle R \rangle\) values are predicted. In Fig. 7, predicted \(\langle R \rangle\) values are plotted as the solid curve, which is in a good agreement with numerical \(\langle R \rangle\) values for \(\tau_c > 2.8\). This result indicates that the heteroclinic cycles become attracting at \(\tau_c \approx 2.8\). When heteroclinic cycles are nonattracting, it can be generally expected that attracting limit-cycles exist close to the heteroclinic cycles. Actually, the phenomenon shown in Fig. 8(d) is rather similar to that in Fig. 8(c), in particular before the system gets very close to two cluster states (e.g., \(t \approx 50\)). Thus, our interpretation of noisy one-cluster state is a noisy dynamics along heteroclinic cycles between unstable, saddle type cluster states.

We also investigate the effect of noise. We consider

\[
\dot{\phi}_i = \omega + \frac{K}{N} \sum_{j=1}^{N} \Gamma(\phi_i - \phi_j) + \sigma \xi_i,
\]

where \(\sigma\) is noise intensity and \(\xi_i(t)\) is white Gaussian noise with zero mean and unit variance. The green triangles in Fig. 7 show \(\langle R \rangle\) values obtained by numerical simulation of Eq. (25). As seen, \(\langle R \rangle\) values are similar to those in the noiseless system for \(\tau_c < 2.8\). Effect of noise is significant \(\tau_c \geq 2.8\) because noise inhibits the system to get very close to unstable two-cluster states [14, 20, 21]. Thus, with noise, the noisy one-cluster state persists even for \(\tau_c \geq 2.8\). By a numerical simulation, we confirmed qualitatively the same result is obtained for nonidentical oscillators (i.e., \(\omega\) in Eq. (25) is \(i\)-dependent). These results indicate that the noisy one-cluster state is robust against noise. Therefore, we interpret that the noisy one-cluster state observed in the experiment is generated by an itinerant synchronization involving unstable, saddle type cluster states.
FIG. 5: Functions in the AIF model. (a) Waveform $\tilde{v}(\phi)$. (b) Phase sensitivity function $Z(\phi)$. (c) Phase interaction functions $\Gamma(\phi)$. $\tau_e = 2.0$ in (a) and (b); and $\tau_e = 0.3$.

FIG. 6: Stability diagram for the AIF system with global coupling. (a) positive coupling. (b) negative coupling. Each circle indicates that the $n$-cluster state is stable at the $\tau_e$ value. Open and filled circles indicate that the largest non-trivial eigenvalue is a real and imaginary value, respectively.

IV. CONCLUDING REMARKS

In summary, we have shown that a noisy synchronized state can occur in a negatively coupled electrochemical oscillator system; this synchronized state was explained theoretically as an itinerant motion among unstable cluster states. This itinerant synchronization could further contribute to the wide range of emergent collective behavior of physical, chemical, and biological oscillators. We expect that similar phenomena can be reproduced in other oscillators when they are close to homoclinic bifurcation. For example, the a Belousov-Zhabotinsky oscillatory reaction also exhibits phase response curve similar to the predictions of AIF model [22]. Neural models are good candidates as well, because many of them, such as the Morris-Lecar model [11], exhibit homoclinic bifurcation.

Many integrate-and-fire models proposed previously assume instantaneous resetting, which yields discontinuous $v(t)$ [12, 15, 21, 23, 24]. In contrast, in the AIF model, we have introduced the resting process with fi-
nite period \( \tau \), which enables us to consider continuous \( \nu(t) \). This feature is not only natural but also a great advantage in mathematical and numerical treatments because then our model has continuous flow at any time. Thereby, delicate problems due to discontinuity can be avoided. For example, it is important that the interaction function \( \Gamma(\phi) \) has continuous derivative at \( \phi = 0 \), i.e., \( \Gamma'(0) = \Gamma'(2\pi) = -\tau \) for nonvanishing \( \tau \), because \( \Gamma'(0) \) plays a vital role in determining many synchronous states.

Acknowledgments

H.K. acknowledges supports from MEXT KAKENHI Grant Number 15H05876. I.Z.K. acknowledges support from National Science Foundation CHE-1465013 grant. One of the authors, H.K., met John Hudson at Fritz Haber Institute in Berlin in 2003, and after that, had many occasions to collaborate with him. All of them were great experiences for H.K., through which H.K learned a lot about how to work on science and enjoy collaborations.

Appendix A: Stability of the balanced cluster states

We briefly summarize the stability analysis of the balanced cluster states [19]. In the phase model given by Eq. (23), the balanced \( n \)-cluster state always exist for any \( \Gamma \). In the balanced \( n \)-cluster state, \( N/n \) oscillators make a point cluster and these oscillators take the same phase \( \Phi_k \) \( (k = 0, 1, \ldots, n-1) \), given by

\[
\Phi_k = \Omega t + \frac{2\pi k}{n},
\]

where \( \Omega \) is the actual frequency. By substituting Eq. (A1) into Eq. (23), we obtain

\[
\Omega = \frac{1}{n} \sum_{k=0}^{n-1} \Gamma \left( \frac{2\pi k}{n} \right).
\]

By solving the eigenvalue problem for the corresponding stability matrix, we obtain \( N \) eigenvalues as

\[
\lambda_1 = \frac{1}{n} \sum_{k=0}^{n-1} \Gamma' \left( \frac{2\pi k}{n} \right),
\]

\[
\lambda_p = \frac{1}{n} \sum_{k=0}^{n-1} \Gamma' \left( \frac{2\pi k}{n} \right) \left( 1 - e^{-2\pi kp/n} \right),
\]

where the former has \( N-n \) multiplicity and is associated with intra-cluster fluctuation; and the latter has 1 multiplicity for each \( p \) \( (p = 0, 1, \ldots, n-1) \) and is associated with inter-cluster fluctuation. There is one trivial eigenvalue \( \lambda_0 = 0 \), which is associated with uniform phase shift. The balanced \( n \)-cluster state is linearly stable if and only if all the remaining eigenvalues have negative real parts.

The derivative of \( \tilde{\Gamma}(\varphi) \) is

\[
\tilde{\Gamma}'(\varphi) = \begin{cases} \tilde{\Gamma}_1(\varphi)/T & \text{for } 0 \leq \varphi \leq \tau, \\ \tilde{\Gamma}_2(\varphi)/T & \text{for } \tau \leq \varphi \leq \tau_e, \\ \tilde{\Gamma}_3(\varphi)/T & \text{for } \tau_e \leq \varphi \leq T, \end{cases}
\]

where

\[
\tilde{\Gamma}_1(\varphi) = \frac{\tau_e}{T} e^{\frac{\varphi}{T}} - \left( -\varphi + \tau_e - \frac{\tau}{T} + 1 \right) e^{-\varphi},
\]

\[
\tilde{\Gamma}_2(\varphi) = -\left( \varphi - \tau + \frac{\tau_e}{T} - 1 \right) e^{T-\varphi} - \left( -\varphi + \tau_e - \frac{\tau}{T} + 1 \right) e^{-\varphi},
\]

\[
\tilde{\Gamma}_3(\varphi) = -\left( \varphi - \tau + \frac{\tau_e}{T} - 1 \right) e^{T-\varphi} - \frac{\tau_e}{T} e^{-\frac{\varphi}{T} (T-\varphi)}.
\]

Note \( \Gamma'(\varphi) = \frac{d\tilde{\Gamma}(\varphi)}{d\varphi} \left( \omega \tilde{\Gamma}(\varphi) \right) = \tilde{\Gamma}' \left( \frac{\varphi}{\omega} \right) \).

Appendix B: Existence and stability of two-cluster states

We briefly summarize the existence and stability analysis of the two-cluster states and the condition for the existence of attracting heteroclinic cycles between a pair of two-cluster states [20, 21]. There is a family of two-cluster states in Eq. (23), in which \( qN \) oscillators and \( (1-q)N \) oscillators form point clusters. Let \( \phi_A \) and \( \phi_B \) be the phase of these clusters. The phase difference \( \Delta \phi = \phi_A - \phi_B \) is obtained as the solution to

\[
(2q-1)\Gamma(0) + (1-q)\Gamma(\Delta \phi) - q\Gamma(-\Delta \phi) = 0.
\]

The eigenvalues of the corresponding stability matrix are

\[
\lambda_1 = Kq\Gamma'(0) + (1-q)\Gamma'(\Delta \phi),
\]

\[
\lambda_2 = K(1-q)\Gamma'(0) + q\Gamma'(-\Delta \phi),
\]

\[
\lambda_3 = K(1-q)\Gamma'(0) + q\Gamma'(\Delta \phi),
\]

with multiplicities \( Nq-1, N(1-q) - 1,1 \), respectively. There is also one trivial eigenvalue \( \lambda_0 = 0 \). Eigenvalues \( \lambda_1 \) and \( \lambda_2 \) are associated with intra-cluster fluctuation, and \( \lambda_3 \) is associated with inter-cluster fluctuation. For generic \( \Gamma \), many of the two-cluster states are saddles; i.e., only a part of eigenvalues are negative. Nevertheless, such saddle states are meaningful because pairs of the two-cluster states form attracting heteroclinic cycles and the system may approach one of them. From here, for simplicity, we only consider two-cluster states with \( q = \frac{1}{2} \). There are a pair of two-cluster states with the phase differences \( \pm \Delta \phi \). The heteroclinic cycle can be formed between this pair of cluster states, if

\[
\lambda_1 > 0, \lambda_2 < 0, \lambda_3 < 0.
\]
Furthermore, the cycle can be attracting, if

\[
\left| \frac{\lambda_1}{\lambda_2} \right| < 1.
\]  

(B6)

The solid line in Fig. 7 is plotted in the following manner. For given \( \Gamma \), Eq. (B1) is solved numerically to find \( \Delta \phi \). Using this \( \Delta \phi \) value, we check Eqs. (B5) and (B6). If both stability conditions are satisfied, we plot a \( R \) value given by Eq. (24) in Fig. 7.

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