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Desynchronization of coupled electrochemical oscillators with pulse stimulations

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Various stimulation desynchronization techniques are explored in a laboratory experiment on electrochemical oscillators, a system that exhibits transient dynamics, heterogeneities, and inherent noise. Stimulation with a short, single pulse applied at a vulnerable phase can effectively desynchronize a cluster. A double pulse method, that can be applied at any phase, can be improved either by adding an extra weak pulse between the original two pulses or by adding noise to the first pulse.

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Synchronization of sets of coupled oscillators occurs in a variety of fields including physics [1], chemistry [2], biology [3–5], neuroscience [6], and medicine [7]. Theories on the synchronization of phase oscillator populations have been developed [2,8,9] and experimentally demonstrated with a population of electrochemical oscillators [10]. However, in some cases synchronization is not desirable. For example, in population biology the increase of migration rates may cause global metapopulation extinction through synchronization [11]. Mutual synchronization among pedestrians walking on a bridge can cause dangerous swaying motion [12]. Pathological synchronization of clusters of neurons are found to be related to tremor activities [13–15].

Stimulation procedures [7], linear [16] (or nonlinear [17]) feedback algorithms, and decrease of interaction strengths [2] are three ways to destroy unwanted synchronization. Among these methods, the easiest to implement in experimental settings is pulse stimulation. Theoretical studies on desynchronization by pulse stimuli have been carried out using phase oscillator models. Depending on the phase, a pulse may either advance or delay the oscillation. Hence, desynchronization can be achieved with a single pulse stimulation of the right intensity and duration by hitting the synchronized system at a vulnerable phase in such a way that approximately half of the elements are delayed, whereas the elements in the other half are advanced [3,7]. Other desynchronization stimulation techniques including double [18] and bipolar double pulses [19] have also been explored with phase models in an attempt to improve the desynchronization effects.

However, experimental studies on desynchronization stimulation techniques are lacking. In this work, we use a synchronized population of electrochemical oscillators that exhibit changes in amplitude, transient dynamics, heterogeneities, inherent noise, and drift as a testbed for various pulse stimulation desynchronization techniques.

The system is an array of 64 nickel electrodes (1-mm diam with 2-mm spacing, in 8×8 geometry) in sulfuric acid. Current, proportional to the rate of reaction (nickel dissolution), is measured on each electrode at a constant applied potential (1.09 V versus K_2SO_4/Hg_2SO_4 reference electrode) just above a Hopf bifurcation; the oscillations are smooth with a mean frequency of 0.485 Hz and a standard deviation of 7 mHz due to heterogeneities of the electrode surface properties. A schematic of the experimental setup and additional experimental details are given in Ref. [10]. Global coupling is added with a series resistor to obtain a synchronized base state of the oscillator population. The phase $\phi_j(t)$ of each element is calculated with the Hilbert transform $[H(i(t)-\langle i\rangle)]$ of individual current time series [10,20]. An order parameter, defined as the normalized length of the vector sum of the phase points $[P_j(t)]$ in $H[i(t)-\langle i\rangle]$ versus $[i(t)-\langle i\rangle]$ space, is used to characterize the extent of the synchrony of the population,

$$Z(t) = \sum_j P_j(t) \Big/ \sum_j |P_j(t)|.$$

This order parameter is similar [21] to the Kuramoto order parameter [2] or the first mode of the generalized order Z_k [9], Z_1. The higher modes of generalized order $Z_k(k>1)$ [7] or clustering algorithms [22] can be used to characterize k cluster states. However, in the experiments on smooth electrochemical oscillators, no clusters were found during the desynchronization.

The magnitude of $r=|Z|$, the order, has a maximum value of 1 for full synchronization and 0 for complete desynchronization (for a population of infinite size). r was adjusted with coupling to be around 0.85–0.95 before the pulse stimulation. The pulse is superimposed on the applied potential and therefore globally affects all elements.

The effect of a single weak pulse administered at a stimulation phase $\phi_S=0.35$ (in fraction of the mean current cycle) is shown in Fig. 1. The amplitude of the mean current [Fig. 1(a)] decreased shortly after the pulse while the individual current reattained its original large amplitude oscillation after two cycles. Simultaneously, the order decreased to a low value [Fig. 1(b)]. Note that r reached the minimum value after a few cycles following the pulse; this is quite different from the effect of a single pulse on phase oscillator populations, where the maximum desynchronization effect is obtained immediately after the pulse. The inset of Fig. 1(b)

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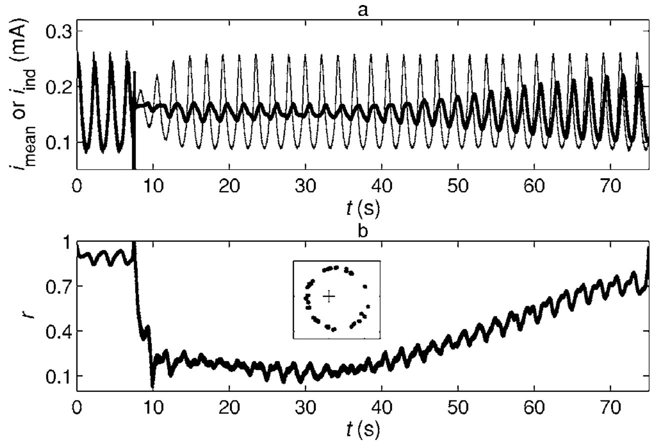

FIG. 1. Suppression of synchrony in population of 64 electrochemical oscillators by a single pulse. The pulse was administered at $t=t_S=7.43$ s, at which the phase of the mean current $\phi_S=\psi_S/(2\pi)$ mod $1=0.35$ (ψ_S is the phase at t_S in radian). Pulse parameters: intensity $I_P=-0.6$ V, duration $T_P=0.1$ s. (a) Time series of the mean (bold line) and an individual (thin line) current. (b) Time series of order r. Inset: Snapshot of oscillators in the $H(i(t)-\langle i\rangle)$ vs $i(t)-\langle i\rangle$ space at $t=34.5$ s.

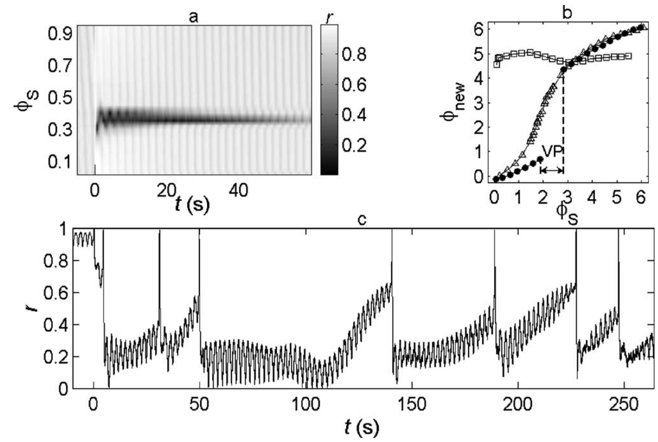

FIG. 2. Effect of single pulse on order parameter. (a) Grayscale plot of r obtained in 20 single pulse experiments in which the pulse was administered at various phases of the mean current (ϕ_S). $I_P=-0.6$ V, $T_P=0.1$ s. The bar on the abscissa indicates the stimulation signal, before which the mean order $r_0=0.89$. (b) Phase of the mean current after the pulse stimulation (ϕ_{new}) vs ϕ_S. Strong (square), weak (circle), and very weak (triangle) pulses with $\{I_P,T_P\}=\{-1\text{ V},0.5\text{ s}\}$, $\{-0.6\text{ V},0.1\text{ s}\}$, $\{-1\text{ V},0.01\text{ s}\}$, respectively. (c) Time series of r in repeated single pulse experiment. The pulse was applied at $\phi_S=0.35$ once the mean current reached an optimized threshold value. The stimulation started at $t=0$ s with $r_0=0.94$, after which the mean order $r_1=0.29$. The average frequency of the pulse administration $f_S=2.7/100$ s.

shows that phase points of the oscillators scattered almost uniformly along the limit cycle in the two-dimensional phase space during the desynchronization state. However, this desynchronized state is unstable and after about 30 s resynchronization began.

The single pulse was administered at various phases of the mean current cycle. The grayscale plot of the order in Fig. 2(a) shows that the single pulse can effectively desynchronize the population only in a narrow range $\phi_S=0.33-0.38$ (5% of the cycle) of the vulnerable phase $\phi_{VP}\cong 0.35$. The value of the vulnerable phase depends on pulse parameters such as intensity and duration. With strong or very weak pulses, only type 0 or type 1 phase resetting, respectively, was obtained as shown in Fig. 2(b). [By definition, type 0 or 1 resetting is characterized by a mean gradient of 0 or 1 in Fig. 2(b).] A vulnerable phase for desynchronization existed at an intermediate pulse strength such that phase singularity [3] occurred [Fig. 2(b)]. We also found that the desynchronization effects are less when the initial order r_0 is large. For example, with $r_0=0.95$, although the vulnerable phase range was still around 0.35, the value of the lowest order after the pulse was more than twice that in the case of $r_0=0.89$ and the resynchronization was about three times faster. Because of the resynchronization, reapplication of the pulse is required to maintain the desynchronized state. A demand-controlled repeated application of the single pulse was implemented in experiments. Figure 2(c) shows a decrease of the mean order from 0.94 to 0.29 in the repeated single pulse operation. There is variability in the series of desynchronization steps. This likely is due to the sensitive dependence of the narrow vulnerable phase range on instantaneous order, pulse parameters, and on minor variations of the system conditions.

One major drawback of the single pulse stimulation is that real-time knowledge of the phase is required to assure that the pulse is triggered in the narrow vulnerable phase range. Phase oscillators have been used to develop double pulse methods in which the first (strong) pulse sets the phase of the cluster to a value ϕ_{PR} independent of its initial dynamical state, the order parameter quickly relaxes back to the original limit cycle, and the second (weak) pulse is applied at a vulnerable phase to desynchronize the cluster [18].

The effect of a strong, phase-reset pulse in the experimental system is shown in Fig. 3(a). Right after the strong pulse, the phase of the order parameter reached a fixed value $\phi_{PR}=0.7$ independent of the stimulation phase [see upper panel in Fig. 3(a)], although some spread of the phase then gradually developed. The order r also increased as a result of the strong pulse [lower panel of Fig. 3(a)] and r slowly relaxed back to its original lower value after more than 28 cycles; this is much slower than that found in simulations with phase oscillators. Correspondingly, a longer relaxation time between the two pulses, on the order of the period of the individual cycle, has to be used to assure that the second, desynchronization pulse will work efficiently. Figure 3(b) shows the effects of the double pulse stimulation with a separation time T_{12}. $T_{12}=\text{Const}\times T_0$, where T_0 is the period of the cycle. (Because of the small drift that exists in the system, T_0 may need to be updated in long experiments.) Const=5.6 is obtained based on the number of cycles n that we waited after the first pulse, the reset phase ϕ_{PR}, and the vulnerable phase ϕ_{VP}, i.e., $\text{Const}=n+(1-\phi_{PR})+\phi_{VP}=5+(1-0.7)+0.3=5.6$. Compared with the single pulse results in Fig. 2(a), we see that the greatest improvement by the double pulse stimulation is that it can desynchronize the cluster irrespective of the stimulation phases, which implies that no real-time phase calculation is required. However, the

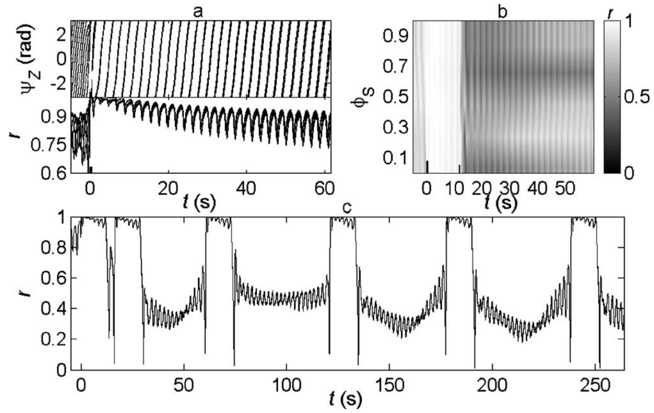

FIG. 3. Desynchronization by double pulse stimulation. (a) Phase reset by a strong pulse ($I_P=-1$ V, $T_P=0.5$ s) in 20 experiments with various stimulation phases. Upper panel: time series of the phase of the order parameter ψ_Z; lower panel: time series of order r. (b) Grayscale plot of r from double pulse experiments. Pulse parameters: first pulse, $I_1=-1$ V, $T_1=0.5$ s; second pulse, $I_2=-0.6$ V, $T_2=0.1$ s; relaxation time between the two pulses, $T_{12}=5.6\times T_0$, where $T_0=2.18$ s is the mean period between the two pulses. (c) Time series of r in repeated double pulse experiment. $r_0=0.89$, $r_1=0.57$, $f_S=2.31/100$ s.

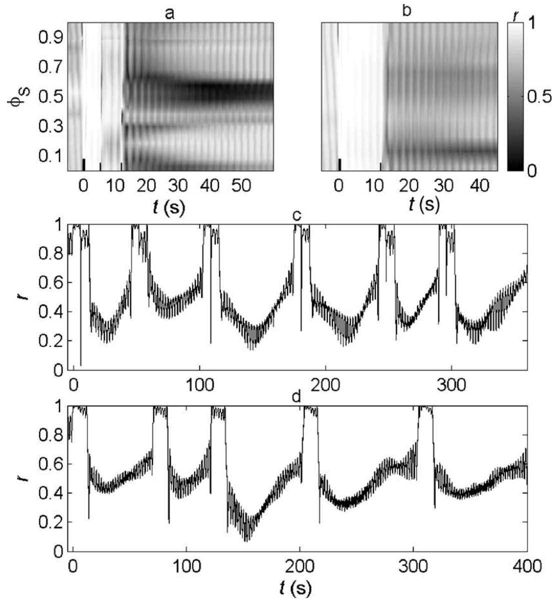

FIG. 4. Desynchronization of the cluster by modified double pulse stimulation. (a) Grayscale plot of order r from triple pulse experiments. Pulse parameters: first pulse, $I_1=-1$ V, $T_1=0.5$ s; second pulse, $I_2=-0.6$ V, $T_2=0.1$ s; $T_{12}=2.55\times T_0$, where $T_0=2.09$ s; third pulse, $I_3=-0.6$ V, $T_3=0.1$ s; $T_{23}=3.1\times T_0$. (b) Grayscale plot of r from noisy double pulse experiments. Pulse parameters: first pulse, $I_1=-1$ V with 30% of white noise, $T_1=0.5$ s; second pulse, $I_2=-0.6$ V, $T_2=0.11$ s; $T_{12}=5.6\times T_0$, where $T_0=2.10$ s. (c) and (d) Time series of r in repeated triple pulse and noisy double pulse experiment, respectively. (c) $r_0=0.89$, $r_1=0.53$, $f_S=1.69/100$ s. (d) $r_0=0.87$, $r_1=0.54$, $f_S=1.25/100$ s.

higher order value before the second pulse resulted in a less effective desynchronization than that in the single pulse method. Repeated administration of double pulses can block the cluster's resynchronization, as shown in Fig. 3(c). Because of the short synchronized windows between the two pulses and lower effectiveness of the second pulse, the mean order decreased to 0.57, almost twice that obtained in the repeated single pulse experiment (0.29).

If the increase in order effected by the phase reset pulse could be eliminated or limited without affecting the reset phase, the desynchronization pulse would be more effective. However, this could not be achieved by a simple decrease in the intensity or duration of the first pulse because a weaker resetting pulse not only gave a smaller order but also did not produce a phase reset. Two other modifications were thus explored in the experiments in order to decrease the high order obtained with the first resetting pulse. In the first one (triple pulse method), we use two pulses instead of one to desynchronize the population after the phase resetting is achieved. The middle pulse is applied close to but not at the vulnerable phase, and it decreased the order by about 10% without changing the phase of the population to a large extent. This yields an efficient third pulse (which is applied at the vulnerable phase) as seen in Fig. 4(a). Note that there is a somewhat larger dependency of the efficiency of the desynchronization pulse on the stimulation phase compared to that of the double pulse stimulation as can be seen by comparing Figs. 4(a) and 3(b); this is likely due to a small phase change produced by the middle pulse. In repeated application of the triple pulse stimulation [Fig. 4(c)] the mean order decreased to 0.53, only 7% lower than that in the repeated double pulse experiments. However, the average frequency of the pulse administration decreased from $f_S=2.31/100$ s in the repeated double pulse case to $f_S=1.69/100$ s in the repeated triple pulse because the third pulse gave not only a lower minimum order value but also a slower resynchronization rate.

In the second modification the decrease of high order after the phase resetting is achieved by modifying the phase resetting pulse. We make use of the fact that noise has a desynchronizing effect [20] and therefore superimpose noise onto the first pulse (noisy double pulse). The grayscale plot of r [Fig. 4(b)] shows the improvement obtained with noise [compare to Fig. 3(b)]. The time series of r in the repeated noisy double pulse experiment [Fig. 4(d)] shows that an even lower pulse frequency $f_S=1.25/100$ s was obtained due to the slower resynchronization rate. The decrease of the pulse administration frequency reduces the ratio of the synchronized to the desynchronized periods; on the other hand, the short synchronized periods between the two pulses are inherent properties of the double pulse methods. To overcome this issue, the stronger resetting pulse can be replaced by a low-frequency pulse train which causes a soft phase reset by entrainment and hence avoids a strong reset being followed by an epoch of synchronization [23].

In summary, we have shown the existence of a phase singularity in a synchronized population of oscillators; this singularity can be used to desynchronize the population without quenching the individual oscillators. We have implemented single and double pulse desynchronization stimulations to an experimental oscillator population; modifications of the methods based on the studies of phase oscillators are needed because of the transient dynamical features of the laboratory system. Short pulses with duration of a few percent of the cycle period and longer relaxation time up to several cycle periods between the two pulses are required in the single and double pulse method, respectively. The single pulse and its repeated administration can effectively suppress the synchrony of the cluster when the phase of the collective signal is calculated onsite to ensure that each pulse is applied within the narrow vulnerable phase range; in addition, the pulse necessary for desynchronization is much weaker than that causing a reset. The double pulse stimulation eliminates the dependence of the desynchronization on the stimulation phases and the need of the phase calculation. However, desynchronization with the double pulse is less efficient due to the slow relaxation of the order after the first pulse. By adding an extra weak pulse between the original two pulses or by adding noise to the first pulse, a significant decrease of the pulse readministration frequency in the demand-controlled repeated application can be attained.

Although the modified pulse stimulation methods were found effective in desynchronization of an electrochemical oscillator population, they could find applications in many systems with similar features such as transient dynamics and inherent noise. More specifically, they can be promising approaches in the improvement of the neurological stimulation aiming at the suppression of pathological, synchronized cerebral activity such as in Parkinson's disease [13,14] or essential tremor [15]. The advantage of using global pulse stimulation is that it may require only a simple modification of the currently used deep brain stimulation technique.

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