Complex Dynamic Structures

DOI: 10.1002/anie.201008194

Engineering of Synchronization and Clustering of a Population of Chaotic Chemical Oscillators**

*Craig G. Rusin, Isao Tokuda, István Z. Kiss, and John L. Hudson**

Dedicated to the Fritz Haber Institute, Berlin, on the occasion of its 100th anniversary

Description, control, and design of weakly interacting dynamical units are challenging tasks that play a central role in many physical, chemical, and biological systems.[1–5] Control of temporal and spatial variations of reaction rates is especially daunting when the dynamical units exhibit deterministic chaotic oscillations that are sensitive to initial conditions and are long-term unpredictable. Control of spatiotemporal chaos by means of suppression of spiral-wave turbulence to standing waves, cluster patterns, and uniform periodic oscillations, in the catalytic CO oxidation on a Pt (110) single-crystal surface has been successfully achieved with linear delayed feedback of the carbon monoxide partial pressure.[6] The same delayed global feedback methodology has also been applied to induce various dynamical clusters in electrochemical corrosion processes[7] and in the light-sensitive Belousov–Zhabotinsky (BZ) reaction.[8] A fundamental problem of tuning the complex structure of chaotic systems is choosing a feedback scheme and appropriate control parameters to steer the system to a desired structure of spatiotemporal reactivity. This objective requires the use of simple yet accurate models of nonlinear processes, their interactions, and responses to external perturbations. When the individual units exhibit periodic oscillatory behavior, the effect of weak feedback and coupling can be described by phase models[4,9] in which the state of each unit is represented by a single variable, the phase of oscillations. Typical phase-synchronized behavior of a population of periodic oscillators is dynamical differentiation (clustering) where the elements form groups (clusters) in which the phases are identical at all times but different from the phases of the elements in the other groups.[9]

Phase models successfully described synchronization and dynamical differentiation (clustering) of oscillatory electrochemical[10] and BZ bead experiments.[11] Experiment-based phase models can also be used for synchronization engineering of periodic oscillators[12] where a carefully designed external feedback is applied to dial-up complex dynamical structures such as stable and sequentially visited dynamical cluster patterns and desynchronization.

Herein, we consider populations of chaotic oscillators that exhibit strong cycle-to-cycle period variations. We show that the feedback scheme can be designed to obtain dynamical synchronization states of phase coherent chaotic oscillators in a quantitative manner. The method relies on the flexibility and versatility of synchronization engineering (originally developed for periodic oscillators[12]) and on the observation that the long-term phase dynamics of chaotic oscillators is often similar to that of noisy periodic oscillators.[13]

An experimental system of uncoupled, phase-coherent, chaotic oscillators was constructed by using an electrochemical cell consisting of 64 Ni working electrodes (99.98 % pure), a Pt mesh counter electrode, and a $Hg/Hg_2SO_4/K_2SO_4$ (saturated) reference electrode, with a 4.5 M H_2SO_4 electrolyte (Figure 1 a) at (11 ± 0.5) °C.[7] A potentiostat (EG&G Princeton Applied Research) was used to set the circuit potential (V_o) of the cell such that the electrodes undergo transpassive dissolution. With a 906 Ω resistor (R_p) attached to each electrode chaotic current oscillations were observed at $V_o = 1.131$ V (Figure 1 b). For each cycle of the chaotic current oscillations (e.g., in Figure 1 b) the peak-to-peak periods were determined with a standard peak-finding algorithm. The observed period distribution of these elements is illustrated in Figure 1 c while the structure of the dynamical attractor can be seen in Figure 1 d. The period distribution of the low-dimensional chaotic attractor is relatively broad and exhibits a multipeak structure, which is characteristic of chaotic behavior obtained from a period-doubling bifurcation route to chaos.[14]

We use order parameters R_1 and R_2 to describe the extent of relative organization of one- and two-cluster states [Eq. (1)],[4,9]

$$R_k = \frac{1}{N}\left|\sum_{j=1}^{N} \exp(\bar{i}k\phi_j)\right| \quad (1)$$

[*] Dr. C. G. Rusin[+]
Department of Chemical Engineering, University of Virginia
Charlottesville, VA 22902 (USA)
Prof. I. Tokuda
Department of Micro System Technology
Ritsumeikan University
Kusatsu, Shiga 525-8577 (Japan)
Prof. I. Z. Kiss
Department of Chemistry, Saint Louis University
St. Louis, MO 63103 (USA)
Prof. J. L. Hudson
Department of Chemical Engineering, University of Virginia
Charlottesville, VA 22902 (USA)
Fax: (+1) 434-982-2658
E-mail: hudson@virginia.edu
[+] Current address: Department of Cardiology
University of Virginia Medical School
Charlottesville, VA 22902 (USA)

[**] This work was supported in part by the National Science Foundation through grant CBET-0730597.

Supporting information for this article is available on the WWW under http://dx.doi.org/10.1002/anie.201008194.

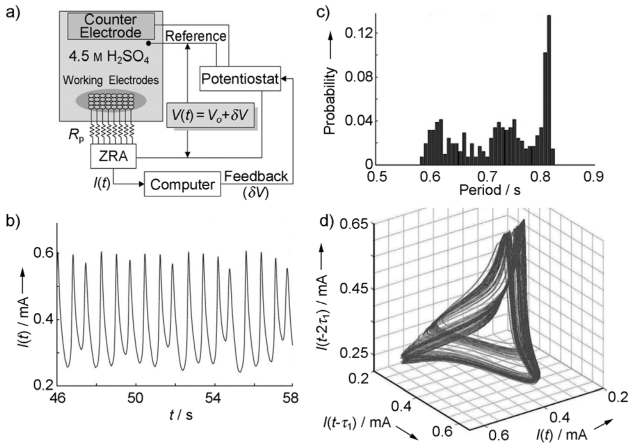

Figure 1. a) Diagram of the experimental apparatus. ZRA: zero-resistance-ammeter box for measuring the currents of the electrodes independently. b) Time-series waveform of the dissolution current of a single nickel electrode. c) Period distribution of a single electrochemical element. d) Reconstruction of the shape of the chaotic attractor using time-delayed embedding, $\tau_1 = 0.224$ s.

where $k = 1,2$, $\bar{i}$ is the imaginary unit, and ϕ_j is the phase of the j-th oscillator obtained with linear interpolation between the current maxima at which the phase is taken to be multiples of 2π.[13] (The value of the order parameters are between zero and one. When a fully synchronized one-cluster state is present, the R_1 and R_2 parameters are about 1; two equally sized cluster states in perfect antiphase configuration would yield a R_1 parameter of about 0 and a R_2 parameter of about 1.) The intrinsic electrical interactions among the oscillators are negligible without external feedback.[10] The phase distribution of the elements was observed to be flat and the R_1 and R_2 order parameters are small (Figure 2b,c) indicating no stable phase cluster states.

Interactions were externally imposed using real-time global feedback of spatially averaged quantities in the form of Equations (2) and (3),

$$h(x(t)) = k_0 + k_S\, x(t-\tau)^S \qquad (2)$$

$$\delta V = \frac{K}{N}\sum_{i=1}^{N} h(x_i(t)) \qquad (3)$$

where δV is the change in the circuit potential, N is the number of oscillators (electrodes), $x_i(t)$ is the scaled dissolution current of element i, K is the overall feedback gain, k_0 and k_S are polynomial feedback coefficients, τ is the feedback time delay, and S is the feedback order. The form of the feedback function was motivated from previous studies[12,15] where it was shown that delayed feedback of the spatial mean of a properly chosen variable raised to the power S has strong impact on the formation of oscillatory dynamical structures with S phase clusters.

A phase model was constructed to describe the effect of feedback on dynamical behavior [Eq. (4)],

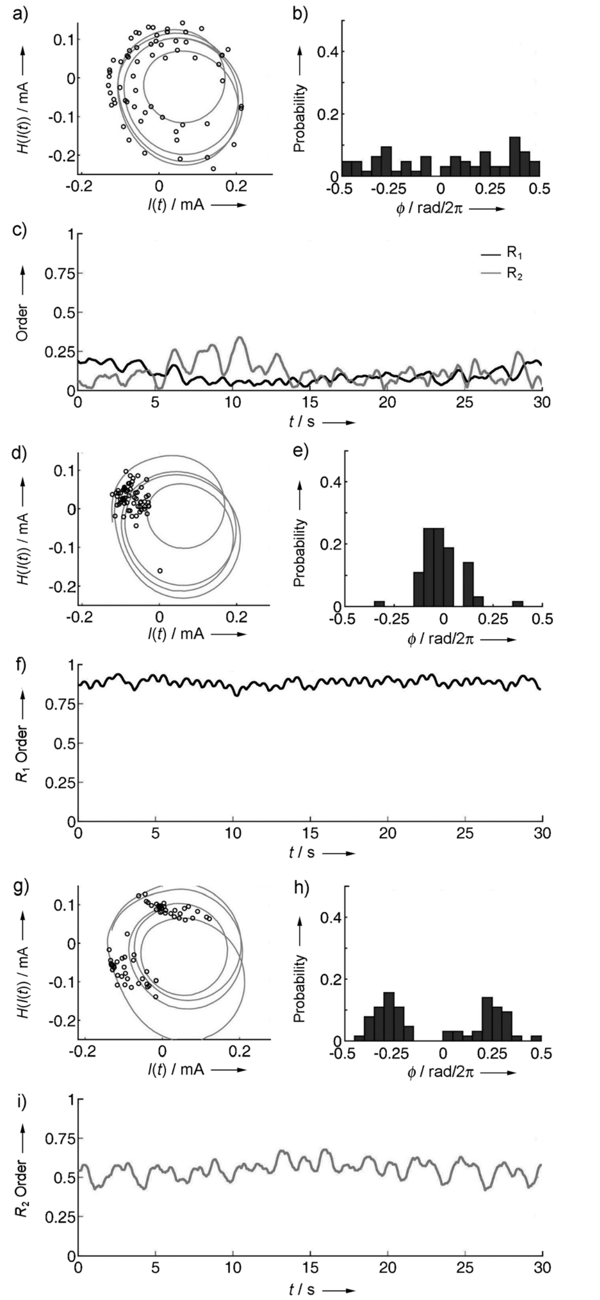

Figure 2. Phase dynamics observed in a population of 64 phase-coherent chaotic electrochemical oscillators without feedback [$K = 0$] (a–c); with linear feedback [$S = 1$, $k_1 = 0.06$, $k_0 = 0$, $K = 1$, $\tau = 0.05$ rad/2π] (d–f); and with second-order feedback [$S = 2$, $k_2 = -0.5$ V^{-1}, $k_0 = -0.007$ V, $K = 1$, $\tau = 0.62$ rad/2π] (g–i). a,d,g) Two-dimensional state–space embedding of the chaotic signal using the Hilbert transform;[13] circles represent the position of the 64 elements in the population, the line is the trajectory of a single representative element over time. b,e,h) Snapshots of the phase distribution of the chaotic elements. c,f,i) Kuramoto order parameters as a function of time for the experimental system.

$$\frac{d\phi_i}{dt} = \omega_i + \frac{K}{N}\sum_{j=1}^{N} H(\phi_j - \phi_i) \qquad i = 1,2,\ldots N \qquad (4)$$

where ϕ_i is the phase of element i, K is the interaction strength, and $H(\Delta\phi)$ is the interaction function. The interaction function can be determined from macroscopic physical quantities [Eq. (5)],

$$H(\Delta\phi) = \int_0^{2\pi} Z(\phi)h(\phi + \Delta\phi)d\phi \quad (5)$$

where $Z(\phi)$ is the response function of the oscillator, and $h(\phi)$ is the physical stimulation of the elements caused by feedback [Eq. (2)].[4] The response function (infinitesimal phase response curve) shows the phase advance per unit perturbation as a function of the phase of the oscillator. The interaction function $H(\Delta\phi)$ is thus obtained as the cycle-average of the phase advance of an oscillator that is induced by an interacting oscillator at a fixed phase difference, $\Delta\phi$. Note also that by virtue of the definition of the amplitude of the interaction function, the overall feedback gain of K in Equation (3) corresponds to an interaction strength K in Equation (4).

The important quantities for the phase model ($H(\Delta\phi)$ and/or $Z(\phi)$) can be directly measured from experiments with a single oscillator ($N=1$); the details are described in the Supporting Information. $H(\Delta\phi)$ can be obtained by recording the average period of the single oscillator as a function of the applied feedback delay. For example, the measured $H(\Delta\phi)$, which represent the interaction functions averaged over many cycles of the chaotic oscillator, are shown in Figure 3 a,b for

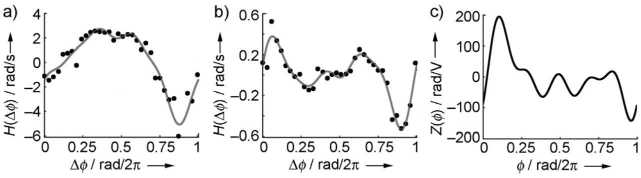

Figure 3. a) Interaction function of a phase-coherent chaotic oscillator under first-order feedback. ($S=1$, $k_1=0.06$, $k_0=0$ V, $K=1$, $\tau=0$ rad/2π) The solid line is the best-fit Fourier approximation. b) Interaction function of a phase-coherent chaotic oscillator under second-order feedback ($S=2$, $k_2=-0.5$ 1/V, $k_0=-0.007$ V, $K=1$, $\tau=0$ rad/2π). c) Response function of a phase-coherent chaotic oscillator.

linear ($S=1$) and quadratic ($S=2$) feedback, respectively. The second-order feedback (Figure 3 b) enhances higher-order harmonics in $H(\Delta\phi)$ more than first-order feedback does (Figure 3 a); this observation is similar to that obtained with periodic oscillators.[12] The response function ($Z(\phi)$) of the phase-coherent chaotic oscillator, shown in Figure 3 c, can then be deconvoluted from Equation (5) using standard Fourier and matrix techniques.[15]

Having obtained the response function of the chaotic oscillator, now we can predict the effect of any type of feedback [e.g., any order and delay in Eq. (2)] by calculating H from Equation (5) with h and Z and then using H in the phase model [Eq. (4)] for analysis of the imposed dynamical response. The typical solution of the phase equations [Eq. (4)] is the occurrence of cluster states where the elements in each cluster synchronize with a common phase. The stability of the observed cluster states can be determined from the eigenvalues associated with a balanced M-cluster state calculated from the Fourier coefficients of the interaction function.[9,16]

The eigenvalue of the balanced one-cluster state (λ_1) was calculated as a function of feedback delay for the experimental system under first-order feedback (Figure 4 a). For a stable cluster state, all of its associated eigenvalues must have negative real parts. The eigenvalues indicate that the exper-

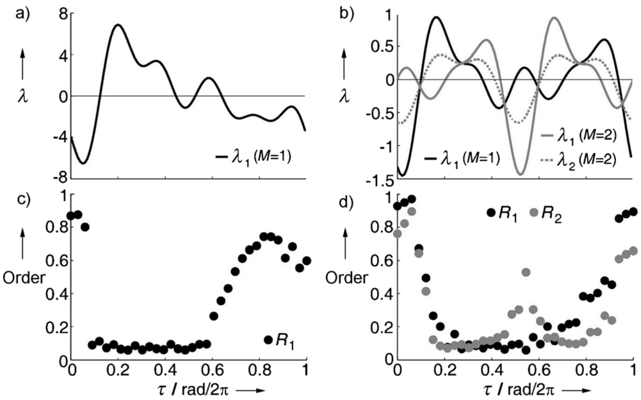

Figure 4. Eigenvalues for the one-cluster ($M=1$) and two-cluster ($M=2$) states of a phase-coherent chaotic system as a function of global feedback delay for a) first-order feedback ($S=1$, $k_1=0.06$, $k_0=0$ V, $K=1$) and b) second-order feedback ($S=2$, $k_2=-0.5$ 1/V, $k_0=-0.007$ V, $K=1$). R_1 and R_2 Kuramoto order parameters for a population of 64 phase-coherent elements under c) first-order and d) second-order feedback.

imental system will transition between a stable one-cluster state and a desynchronized state at a feedback delay of approximately 0.15 rad/2π. Additionally, the experimental system will transition from a desynchronized state to a stable one-cluster state at a feedback delay value of approximately 0.61 rad/2π.

Indeed, as it is shown in Figure 2 d,e, upon application of first-order feedback stimulation with $\tau=0.05$ rad/2π, the phases of the elements were observed to synchronize into a single-phase cluster state while the trajectories of the individual elements were observed to be chaotic. The R_1 order parameter was calculated from the experimental data and had a value of approximately 0.87 for the duration of the experiment, indicating a stable one-cluster state (Figure 2 f). The order parameters calculated from the experimental data as a function of feedback delay can be seen in Figure 4 c. Under first-order feedback stimulation, a high R_1 order parameter was observed between $[0 \leq \tau \leq 0.07]$ rad/2π as well as $\tau > 0.6$ rad/2π indicating the presence of a balanced one-cluster state.

The cluster stability calculation was repeated for the experimental system by using second-order global time-delayed feedback stimulation. The eigenvalues for the one-cluster and the two-cluster states were determined as a function of the feedback delay (Figure 4 b). The eigenvalues indicate a stable one-cluster state when the feedback delay is approximately 0 or 1 rad/2π, as well as a stable two-cluster state when the feedback delay is between 0.45 and 0.6 rad/2π.

Upon application of second-order feedback with $\tau = 0.62$ rad/2π to a population of oscillators, a two-cluster state was observed (Figure 2 g–i). The phase distribution and R_2 order parameters indicate the presence of a stable two-cluster state. Although the phases of the individual elements were synchronized, their trajectories remained chaotic. (Note that these mild feedback-induced phase clusters differ from the chaotic clusters obtained with strong feedback and coupling[7] in that the elements in the same cluster are phase- but not identically synchronized.)

The order parameters calculated from the experimental data as a function of feedback delay for quadratic feedback can be seen in Figure 4 d. A high R_1 order parameter was observed between $[0 \leq \tau \leq 0.07]$ rad/2π and $[0.90 \leq \tau \leq 17]$ rad/2π indicating the presence of a balanced one-cluster state. Additionally, a high R_2 order parameter was observed for $[0.5 \leq \tau \leq 0.6]$ rad/2π indicating the presence of a balanced two-cluster state.

Comparison of the phase model predictions for the eigenvalues in Figure 4 a,b to the experimental observations in Figure 4 c,d indicates that the phase model accurately predicts the location of both the one-cluster and two-cluster states. Synchronized cluster states are not experimentally observed where the eigenvalues indicate the lack of any stable cluster state.

We have also carried out numerical simulations with a pair of chaotic Rössler oscillators with global delayed feedback. (Details are given in the Supporting Information.) The numerical simulations again show the applicability of the phase-model-based approach for the description and design of various synchronized states with global feedback. The results indicate that the interaction function of the chaotic behavior is very similar to that of the periodic orbits close to the chaotic behavior in parameter space. In general, chaotic phase synchronization is based upon temporal synchronization of unstable periodic orbits embedded in the chaotic attractor;[13,17] the results imply that chaotic phase synchronization may be described by stochastic switching of very similar phase equations of the various unstable periodic orbits embedded in the chaotic attractor. This would allow the phase model, with a single interaction function estimated by the delayed feedback technique, to accurately describe the collective dynamics for systems of phase-coherent chaotic elements.

Our experimental and numerical findings indicate that phase-coherent chaotic oscillator populations that exhibit strong cycle-to-cycle variations of periods and amplitudes can be steered towards designed complex synchronization structures with global delayed feedback of spatially averaged quantities. The proposed method extracts a fundamental oscillator property, the response function, from measurements of the mean period and the waveform of a single chaotic oscillator with external feedback. The response function then can be applied to predict quantitatively the effect of nonlinear, delayed global feedback on the synchronization structure of a chaotic oscillator population. Although the method was demonstrated for one- and two-cluster states, the general approach can be applied to many cluster states using higher-order feedback[15] as well as to destroying emergent dynamical structures, for example, desynchronization.[12] The robustness of the methodology to dynamical jitter of the oscillations makes it a prospect for applications to control firing patterns of small neuron circuitries as well as tuning spatiotemporal cortical dynamics.[18]

Received: December 26, 2010
Published online: April 6, 2011

Keywords: chaos · electrochemistry · nonequilibrium processes · phase models · synchronization

[1] G. Ertl, *Science* **1991**, *254*, 1750.
[2] A. S. Mikhailov, K. Showalter, *Phys. Rep.* **2006**, *425*, 79.
[3] V. S. Zykov, H. Engel, *Physica D* **2004**, *199*, 243–263.
[4] Y. Kuramoto, *Chemical Oscillations, Waves, and Turbulence*, Springer, New York, **1984**.
[5] I. Z. Kiss, J. L. Hudson, *AIChE J.* **2003**, *49*, 2234–2241.
[6] M. Kim, M. Bertram, M. Pollman, A. von Oertzen, A. S. Mikhailov, H. H. Rotermund, G. Ertl, *Science* **2001**, *292*, 1357.
[7] W. Wang, I. Z. Kiss, J. L. Hudson, *Phys. Rev. Lett.* **2001**, *86*, 4954–4957.
[8] V. K. Vanag, L. F. Yang, M. Dolnik, A. M. Zhabotinsky, I. R. Epstein, *Nature* **2000**, *406*, 389–391.
[9] S. C. Manrubia, A. S. Mikhailov, D. H. Zanette, *Emergence of Dynamical Order: Synhronization Phenomena in Complex Systems*, World Scientific, Singapore, **2004**.
[10] I. Z. Kiss, Y. M. Zhai, J. L. Hudson, *Phys. Rev. Lett.* **2005**, *94*, 248301.
[11] A. F. Taylor, P. Kapetanopoulos, B. J. Whitaker, R. Tóth, L. Bull, M. R. Tinsley, *Phys. Rev. Lett.* **2008**, *100*, 214101.
[12] I. Z. Kiss, C. G. Rusin, H. Kori, J. L. Hudson, *Science* **2007**, *316*, 1886–1889.
[13] A. Pikovsky, M. Rosenblum, J. Kurths, *Synchronization: A Universal Concept of Nonlinear Sciences*, Cambridge University Press, Cambridge, **2001**.
[14] J. Davidsen, I. Z. Kiss, J. L. Hudson, R. Kapral, *Phys. Rev. E* **2003**, *68*, 026217.
[15] H. Kori, C. G. Rusin, I. Z. Kiss, J. L. Hudson, *Chaos* **2008**, *18*, 026111.
[16] K. Okuda, *Physica D* **1993**, *63*, 424.
[17] A. Pikovsky, M. Zaks, M. Rosenblum, G. Osipov, J. Kurths, *Chaos* **1997**, *7*, 680–687.
[18] O. V. Popovych, C. Hauptmann, P. A. Tass, *Biol. Cybern.* **2006**, *95*, 69.