Synchronization and Clustering of Arrays of Electrochemical Oscillators with Global Feedback

Wen Wang, István Z. Kiss, and John L. Hudson*

Department of Chemical Engineering, University of Virginia, 102 Engineers' Way, Charlottesville, Virginia 22904-4741

Experiments on chaotically oscillating arrays of nickel electrodes in sulfuric acid were carried out. A global feedback is added in which a signal proportional to the difference between the sum of the currents of all elements and a mean current is fed back to the applied potential. The addition of global feedback transforms a system of weakly coupled elements to one of complete synchronization. At intermediate feedback strengths, intermittent, unstable chaotic clusters are observed in which clusters of elements form and break up. Stable clusters or condensates form at somewhat higher feedback strengths. The stable clusters become periodic at higher gain; both two-cluster and three-cluster states are observed. With a further increase in the feedback gain, one of the clusters dominates the collective dynamics; its size increases until at some feedback strength the entire system is synchronized. Delay in the feedback signal also influences the interaction of the oscillators.

Introduction

The behavior or collective dynamics of a large set of oscillating elements depends both on the characteristics of each individual entity and on the type and degree of coupling among the sites. When no coupling is present, the entities all act independently of the others. Sufficiently strong coupling can synchronize the elements. At intermediate coupling strengths the elements influence each other, although synchronization is not attained. Under some conditions, clustering or condensation occurs in which elements in subsets undergo the same behavior.[1]

The interaction of sets of nonlinear oscillators at various time and space scales has importance in a number of fields such as communications,[2] population dynamics, neural systems,[3,4] and distributed chemical reaction systems.[5] In each case, several individual sites, at which, for example, chemical, biological, or electronic processes occur, are coupled so that the sites interact. The examples differ, of course, both in the nature of the individual sites and in the mechanism of the coupling. Nevertheless, similar trends in collective dynamics can often be observed. In population biology the individual units are entire organisms and the interactions can arise through competition for resources.[6] The growth and spread of viral diseases within a human population are also influenced by the interactions among the individuals.[7] In neurobiology, the units are neurons or groups of neurons and the interactions arise through a network of axons and dendrites; one question here is the tendency of groups of neurons to oscillate in a regular, nonchaotic fashion, while the individual neurons without coupling are chaotic.[8] In communications, the synchronization of two chaotic circuits can be potentially used to transmit signals.[2] In chemically reacting systems, the individual entities are reaction sites and the coupling is through diffusion or convection or, as in the case of electrochemical reactions, also through the electric field.[5,9−11]

Although theoretical studies using models derived from chemistry,[12,13] biology,[14−17] and physics[1,2,18−23] have explored the emergence of synchronized and clustered states due to coupling, laboratory studies are rare. We present here such an experimental study carried out with an apparatus consisting of an array of chemical oscillators (electrodes) and mechanisms of independently changing the coupling among the elements. The use of discrete elements is in contrast to some previous experimental studies in which global coupling[24] as well as forcing and feedback[25−28] was applied to continuous distributed chemical reaction systems. The specific system being investigated is the electrodissolution of nickel in sulfuric acid.[29] Because the current of each of the electrodes can be measured independently, the experimental system can be used to explore the behavior of coupled systems that cannot be seen in experiments in other types of systems. In addition, an array of discrete electrodes has been shown to behave the same as a single, larger continuous electrode (at length scales larger than that of the size of the individual electrodes),[5] and thus the experiments can be used to study spatiotemporal dynamics of electrochemically reacting systems.[11]

The experiments are carried out with the application of global feedback. Proportional control is used in which a signal, based on the difference between the total current (or equivalently the sum of the currents of the individual electrodes) and a mean current, is fed back to the applied potential. Similar feedback has been used to stabilize unstable steady states in nondistributed systems.[30−33] Proportional feedback has also been applied in theoretical studies of chaotic distributed systems, viz., to the complex Ginzburg−Landau equation [34] and to a model of carbon monoxide oxidation.[35] In both cases the suppression of spatiotemporal turbulence was observed. The feedback has two effects in spatiotemporal systems: it supplies a global coupling in which the feedback influences all of the elements equally, and it also affects the dynamics of the individual oscillators.

* Corresponding author. E-mail: hudson@virginia.edu.

10.1021/ie0100737 CCC: $22.00
Published on Web 05/04/2001

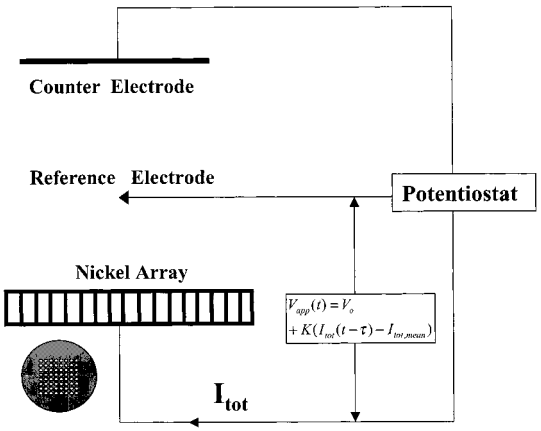

Figure 1. Schematic of the apparatus with feedback.

In this work we carry out an experimental study in which global feedback is applied to an array of chaotic oscillators. We show how changes in the feedback gain K and the feedback time delay influence the interaction of the oscillators; transitions to clustered or condensed states and to synchronized states are investigated.

Experimental Section

The experiments were carried out in a conventional three-compartment electrochemical cell. The schematic of the apparatus including a 64-electrode array working electrode is shown in Figure 1. We also used an 8-electrode array made up of one row of the 64-electrode array in studies of the effects of delay time. The potentials of all of the electrodes in the array are held at the same value relative to a $Hg/Hg_2SO_4/K_2SO_4$ reference electrode with a potentiostat (EG&G Princeton Applied Research, model 273). The counter electrode is Pt mesh. The electrodes are connected to the potentiostat through individual parallel resistors and through one collective series resistor. The zero resistance ammeter (ZRA) circuitry was inserted between the individual and collective resistors to measure the current of each electrode. The voltage from the ZRAs was digitized using a transputer (ADWin Pro, Keithley). The transputer has its own CPU and memory, which ensure fast data acquisition and real-time response. The data acquisition frequency is 100 Hz for all data shown. The calculated feedback signal is superimposed onto the constant potential (V_0) through the analogue output of the transputer. Testpoint software was used to visualize and save the experimental data on a PC.

Experiments were carried out in a 4.5 M H_2SO_4 solution directly diluted from 5 M sulfuric acid. The cell was held at 11 °C.

We have developed a method of altering the strength of global coupling while holding other parameters constant. It is achieved by holding the total resistance inserted into the circuit constant and changing the global coupling parameter ϵ, which is the ratio of collective resistance R_{coll} and total resistance R_{tot}. Further details can be found in a previous paper.[36,37] All of the experiments in the present paper were done at a fixed value (0.56) of the global coupling parameter.

The real-time total current is obtained by summing the individual currents, i.e., those obtained from the ZRA circuitry. A feedback signal is then generated through the D/A system using the specified gain (K) and delay (τ). The perturbation of output voltage is proportional to the difference of the total current $[I_{tot}(t - \tau)]$ and the precalculated mean total current $I_{tot,mean}$:

$$\delta V(t) = K[I_{tot}(t - \tau) - I_{tot,mean}] \quad (1)$$

The applied potential is then

$$V_{app}(t) = V_0 + K[I_{tot}(t - \tau) - I_{tot,mean}] \quad (2)$$

Because the feedback is a small-amplitude perturbation to the applied potential, we assume that the mean total current does not change significantly after the feedback is imposed. The mean total current ($I_{tot,mean}$) is used as the offset of the feedback signal to maintain the applied potential in the chaotic region. For all experiments, a mean individual current was precalculated by integrating the current from one electrode over 20 s before the feedback is applied. The mean total current ($I_{tot,mean}$) was then determined by multiplying the mean individual current by the total number of electrodes on the array.

Results

A. Array without Feedback. Some results on the 64-electrode array without the imposition of feedback are shown in Figure 2. The currents of all 64 electrodes are shown as a space/time gray scale plot in Figure 2a; the current from a single electrode and an attractor in state space constructed from it are shown in parts b and c of Figure 2, respectively.

As indicated in the time series and the attractor, the behavior of each electrode is chaotic. (The time series and attractors of the other 63 electrodes are similar.) It can be seen from the space/time plot that the elements are not synchronized under these conditions, but rather each electrode has a behavior that is somewhat different from that of the others. Nevertheless, the state is not fully turbulent, and there is some order among the elements. The behavior was obtained by adding some global coupling to the array through the resistors. A complete description of the effect of the added global coupling can be found in an earlier publication.[37]

The behavior seen in Figure 2 is used as a base state for the experiments to be described here; i.e., the feedback is added to it. We use this as the base state, rather than a less ordered state, for two reasons. First, the feedback is not strong enough to synchronize the elements without any added global (resistive) coupling under the conditions of these experiments. Second, an objective of the experiments is to explore the formation of clusters and the transition into synchronized states, and so a state closer to a clustered state is chosen as the starting point.

B. Effect of Added Global Feedback. We investigate now the effect of the addition of a global feedback to the behavior shown in Figure 2. After the imposition of the feedback, the system goes through a transient before reaching a final, stationary state. We describe here first the stationary state; the transient is discussed briefly below.

Results obtained at six values of the feedback gain, with all other parameters held constant, are presented in Figures 3−5. Figure 3 shows space/time plots made from the time series of all 64 electrodes in the array; the current is represented in gray scale, with dark shading corresponding to a high current. Some selected time series of individual electrodes and some differences

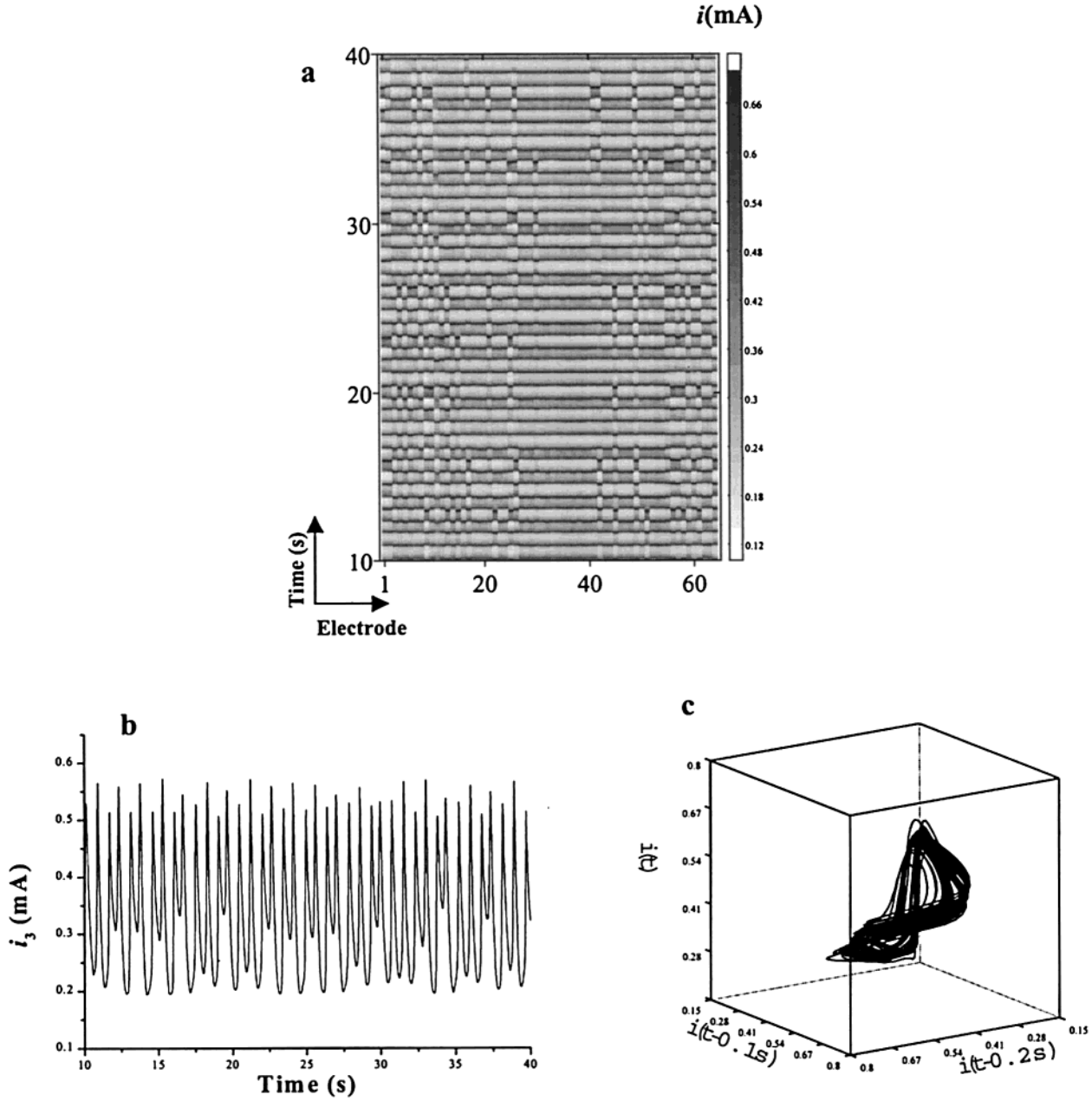

Figure 2. Behavior on a 64-electrode array without feedback ($K = 0$, $\epsilon = 0.56$, $V_{app} = 1.35$ V, $R_{eq} = nR_{tot} = 906\ \Omega$, $\tau = 0$ s): (a) space/time plot of individual currents; (b) time series of individual current; (c) attractor from current on electrode 3.

between pairs of electrodes are shown in Figure 4. In Figure 5 some attractors made from the time series are shown.

We note again that the behavior obtained without feedback, i.e., for $K = 0$, can be seen in Figure 2 for comparison.

Behavior obtained with a low feedback gain ($K = 1.2$ mV/mA) is seen in Figures 3a, 4a, and 5a. Even at this low value, some tendency for synchronization is observed. Some synchronization occurs during short time periods, for example, at $t = 46$ and 68 s, but these synchronized states are not stable and none of them lasted for more than 3 s (approximately four oscillations). Some clusters also tend to form, but again these break up after a short time. Thus, the feedback is too weak to stabilize any ordered structure. The difference between the currents of two electrodes (6 and 7) is shown in Figure 4a. One sees the forming and breaking up of clusters between the two electrodes as the current difference becomes small and then larger, respectively. The attractor shown in Figure 5a is very similar to the one presented in Figure 2c, indicating that the dynamics of the individual oscillators are not altered by the addition of this weak feedback signal.

Intermittent Chaotic Clusters. As we increase the feedback gain (K) to 1.5 mV/mA (Figure 3b), intermittent clusters are observed. These clusters (still unstable) exist for longer times than those occurring at lower values of K, as can be seen by comparison of parts a and b of Figure 4; the elements exist in clusters for many more oscillations at the higher value of K. Nevertheless, there is no sharp demarcation between the behaviors of parts a and b of Figure 4. It can be seen in Figure 4b that the formation of unstable clusters occurs with different elements. That is, say that some (transient) clusters exist on the array such as that occurring up to time approximately equal to 70 s in Figure 4b. Electrodes 45 and 47−49 (and others, of

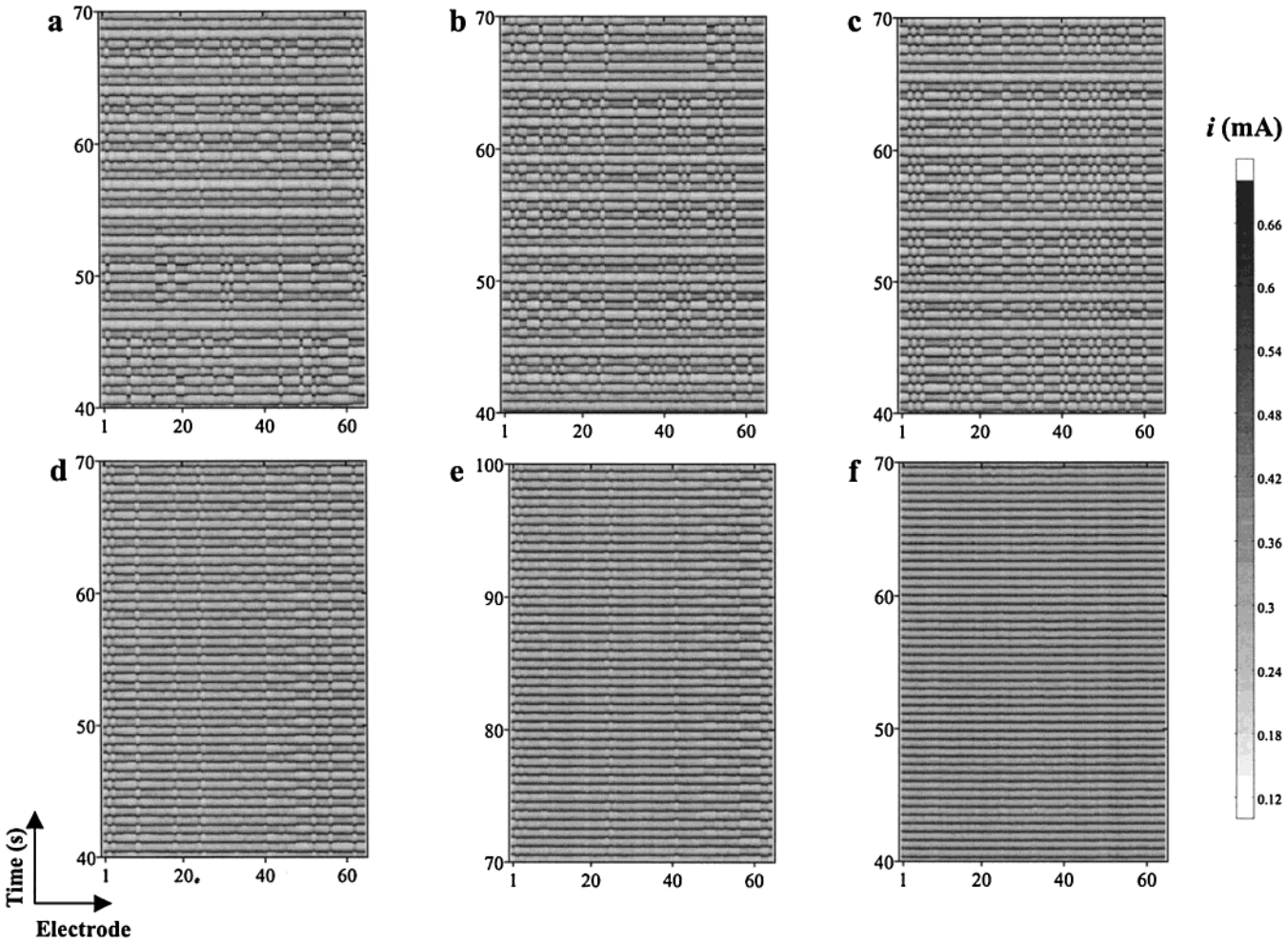

Figure 3. Space/time plots of individual currents (64-electrode array, $\epsilon = 0.56$, $V_0 = 1.35$ V, $R_{eq} = 906\ \Omega$, $\tau = 0$): (a) $K = 1.2$ mV/mA; (b) intermittent clusters, $K = 1.5$ mV/mA; (c) stable chaotic clusters, $K = 2.2$ mV/mA; (d) stable periodic clusters, $K = 2.8$ mV/mA; (e) stable periodic clusters, $K = 3.2$ mV/mA; (f) stable periodic synchronized state, $K = 3.8$ mV/mA.

course) are in one cluster. We show the differences in currents between the two pairs 45 and 47 and 48 and 49. During the time that the transient cluster exists, up to $t = 70$ s, each of these differences is small. At $t = 70$ s the cluster breaks apart, but another cluster forms approximately 7 s later. Electrodes 45, 47, and 49 all are in this new cluster, but 48 is not. This can be seen in the two time series showing differences in currents. This behavior continues. Clusters are formed and break up, and then different clusters are formed. We also note that when a cluster is formed, the individual currents exhibit periodic-like behavior consisting of well-distinguishable large- and small-amplitude oscillations. This can be seen in the time series of the current of electrode 45 at times between 53 and 65 s. Finally we note that at this feedback gain the dynamics of the individual oscillators do not differ significantly from that obtained without feedback; the somewhat distorted attractor (Figure 5b) resembles those seen in Figures 2c ($K = 0$) and 5a ($K = 1.2$ mV/mA).

Stable Chaotic Clusters. Two stable chaotic clusters are formed when the feedback gain is increased to higher than 1.8 mV/mA. An example, obtained at $K = 2.2$ mV/mA, is shown in Figures 3c, 4c, and 5c. The cluster configuration of Figure 3c is (29, 35); that is, there are 29 elements in one cluster and 35 in the other. All of the elements in a given cluster have the same dynamics; their time series are almost identical and differ only slightly because of noise and heterogeneities. The (29, 35) cluster arrangement is shown schematically in Figure 6a; the black elements are in one cluster the white in the other. These cluster arrangements are stable; the cluster arrangement stays as shown throughout the entire experiment. It is also stable to small perturbations. The individual current of each cluster (Figure 4c) is chaotic. The banded feature of the attractor typical for clusters can be seen in Figure 5c. The current difference between the two electrodes in the same clusters (electrodes 1 and 2) is small and remains so throughout the experiment because the cluster is stable and its elements never deviate from the cluster. If two electrodes are in different clusters, such as electrodes 2 and 3, the current difference between them fluctuates with the same magnitude as their individual time series. The cluster arrangement shown in Figures 3c and 6a is only one of a large number that can be obtained at these conditions. We show one example of a (29, 35) cluster. The arrangement obtained depends on the initial conditions, and because the number of possible states is large, the same final stationary state is never seen twice. For example, numbers of elements in the two clusters need not be 29 and 35. Furthermore, even if a (29, 35) state is obtained, the particular elements that lie in the 29 cluster and the 35 cluster differ. More examples of such cluster formations can be found in a previous paper on global coupling.[37]

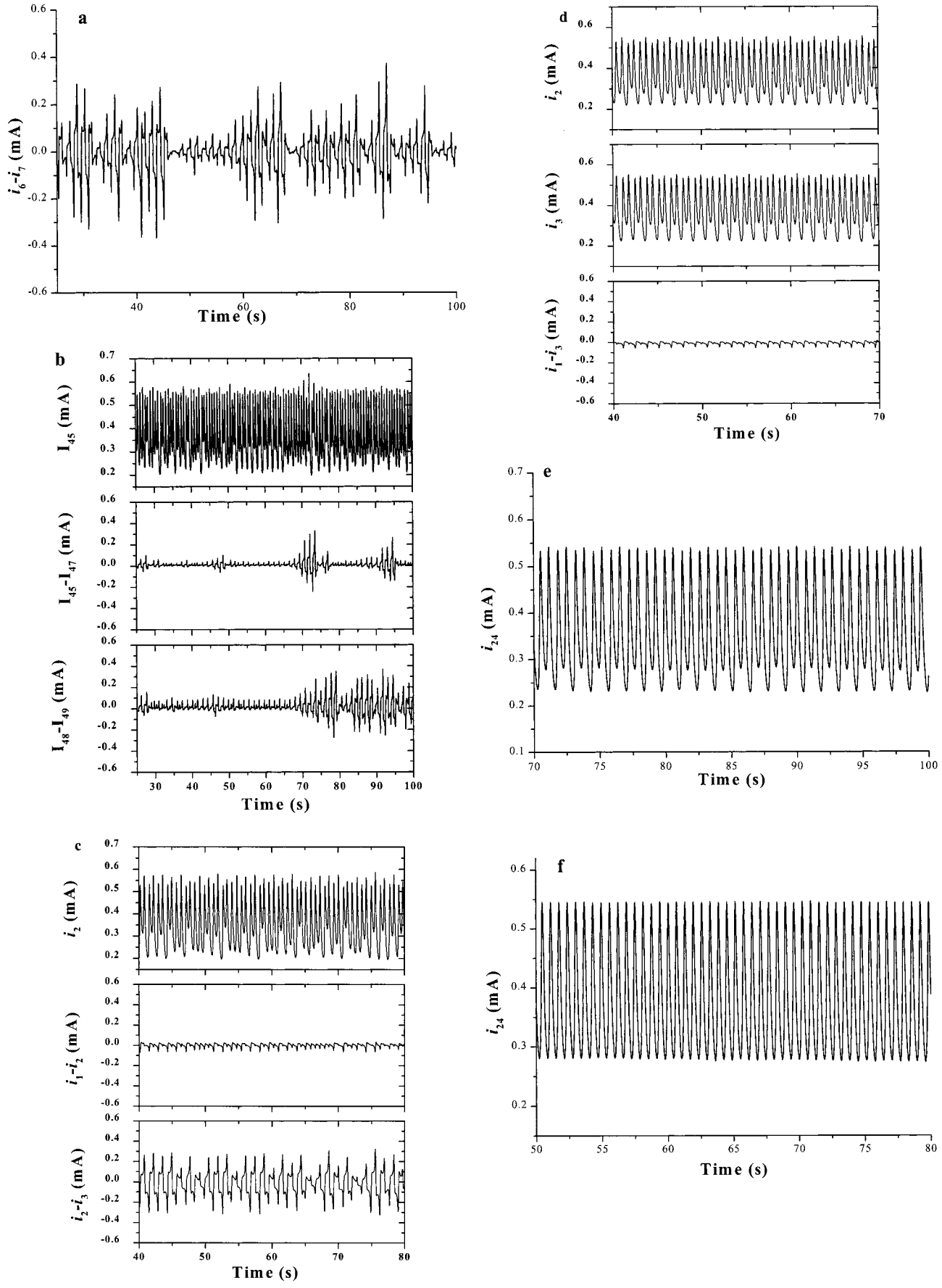

Figure 4. Time series of individual electrodes and differences between pairs (64-electrode array, $\epsilon = 0.56$, $V_0 = 1.35$ V, $R_{eq} = 906\ \Omega$, $\tau = 0$): (a) $K = 1.2$ mV/mA; (b) intermittent clusters, $K = 1.5$ mV/mA; (c) stable chaotic clusters, $K = 2.2$ mV/mA; (d) stable periodic clusters, $K = 2.8$ mV/mA; (e) stable periodic clusters, $K = 3.2$ mV/mA; (f) stable periodic synchronized state, $K = 3.8$ mV/mA.

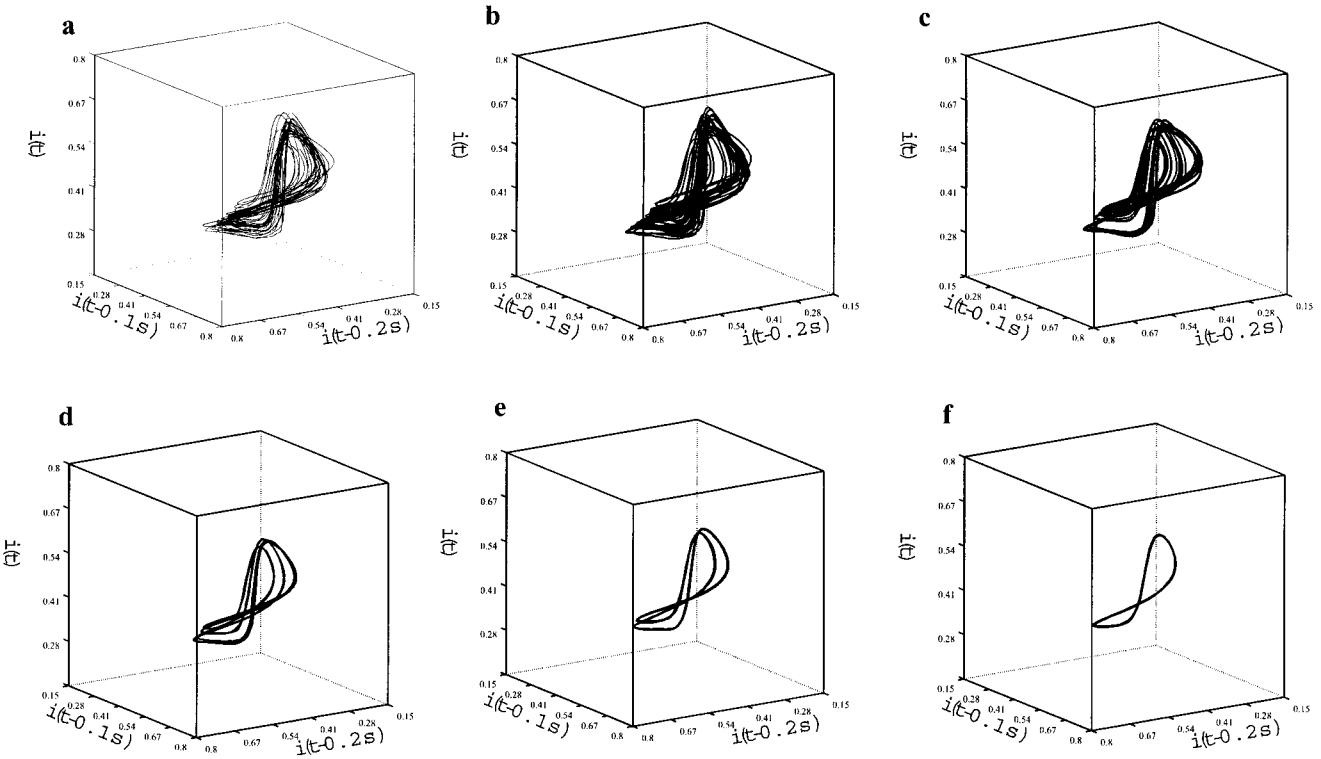

Figure 5. Attractors corresponding to Figures 3 and 4 (64-electrode array, $\epsilon = 0.56$, $V_0 = 1.35$ V, $R_{eq} = 906\ \Omega$, $\tau = 0$): (a) electrode 2, $K = 1.2$ mV/mA; (b) electrode 3, intermittent clusters, $K = 1.5$ mV/mA; (c) electrode 4, stable chaotic clusters, $K = 2.2$ mV/mA; (d) electrode 2, stable periodic clusters, $K = 2.8$ mV/mA; (e) electrode 24, stable periodic clusters, $K = 3.2$ mV/mA; (f) electrode 3, stable periodic synchronized state, $K = 3.8$ mV/mA.

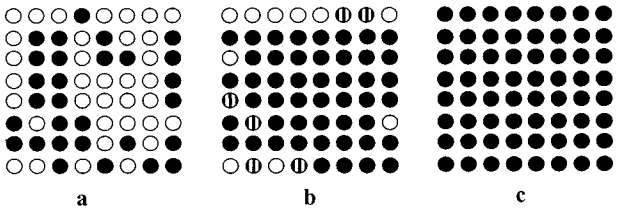

Figure 6. Representative stable cluster arrangements (64-electrode array, $\epsilon = 0.56$, $V_0 = 1.35$ V, $R_{eq} = 906\ \Omega$, $\tau = 0$): (a) two (29, 35) chaotic clusters, $K = 2.2$ mV/mA; (b) three (6, 10, 48) periodic clusters, $K = 3.2$ mV/m; (c) synchronized periodic state, $K = 3.8$ mV/mA.

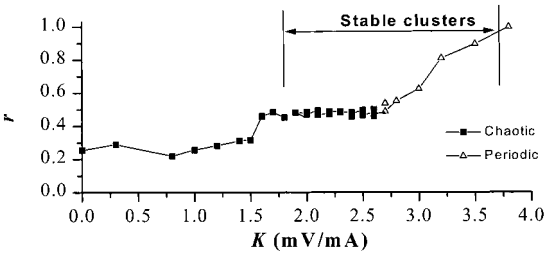

Figure 7. Mean order parameter as a function of feedback gain (64-electrode array, $\epsilon = 0.56$, $V_0 = 1.35$ V, $R_{eq} = 906\ \Omega$, $\tau = 0$).

Stable Periodic Clusters. The stable chaotic clusters changed to stable periodic clusters as we further increased the feedback gain. An example of a periodic two-cluster configuration obtained at $K = 2.8$ mV/mA is shown in Figure 3d. The periodic behavior for both clusters is period 4, as can be seen in the time series (Figure 4d) and in the attractor (Figure 5d). The small value of current difference between electrodes 1 and 3 is an indication of the stability of the clusters.

Another example of stable periodic clusters, obtained at a somewhat higher feedback gain, is shown in Figures 3e, 4e, and 5e. Three clusters occur as seen in Figure 6b with a configuration (6, 10, 48). The time series for the two smaller clusters are period 2 as shown in Figures 4e and 5e. The large cluster containing 48 elements is period 1 to within the accuracy of the experiment. Again, other cluster arrangements are possible at these conditions although their characteristics are similar to those shown here in this example.

Higher Feedback Gains. As the gain is increased, a state with synchronized period 1 oscillations is obtained (Figures 3f, 4f, and 5f). With additional increases, a stable steady state occurs.

Trends in Cluster Configurations. Representative cluster arrangements are shown in Figure 6 for three values of the feedback gain. As noted above, these are only representative, and many configurations occur at any given value of the feedback gain; the specific cluster configuration obtained depends on the initial conditions. However, some trends can be seen. In the chaotic region two clusters are always obtained, and the numbers of elements in the two clusters are approximately evenly balanced. The distribution ranged from (32, 32) to (28, 36); one of the (29, 35) configurations is shown in (Figure 6a). As the feedback gain is increased and the behavior becomes periodic, two trends are noticed in the configurations: an imbalance in cluster sizes occurs, and a third cluster arises. As the feedback gain is further increased, one of the clusters begins to dominate, as seen in Figure 6b, and with a further increase, this cluster encompasses the entire array and synchronization is obtained.

Order Parameter. We have calculated the average pair distances in three-dimensional state space (Figure 5) between each electrode pair of the 64 elements as a function of time. An order parameter can be defined as a fraction of the number of pairs whose distance in the

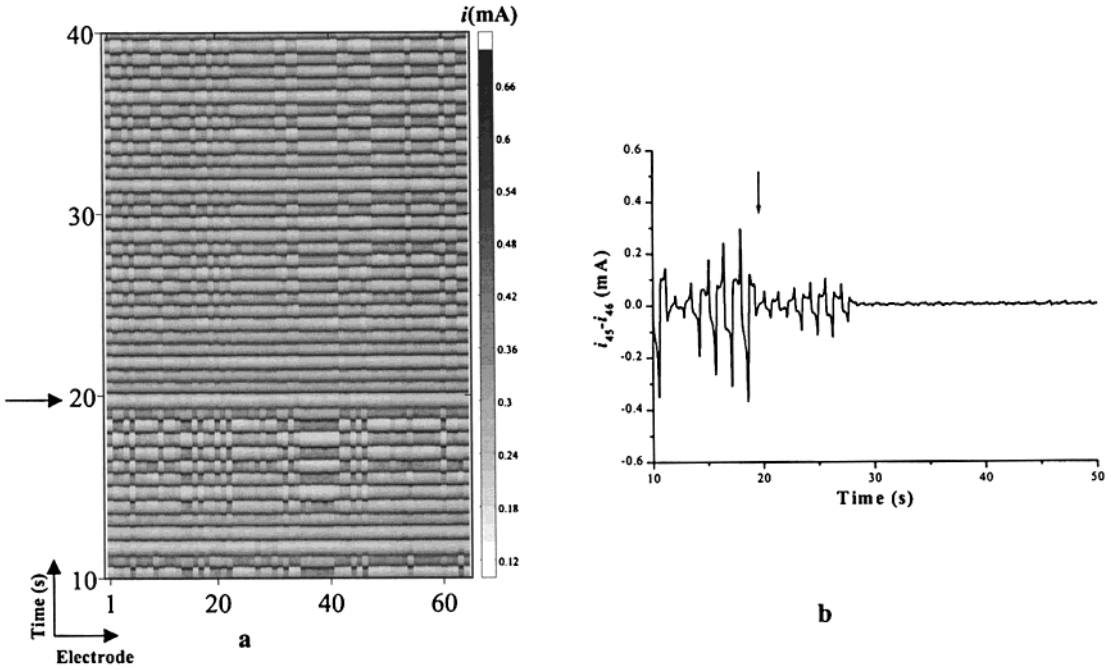

Figure 8. Initial transient leading to stable chaotic clusters (64-electrode array, $\epsilon = 0.56$, $V_0 = 1.35$ V, $R_{eq} = 906\ \Omega$, $K = 2.2$ mV/mA, $\tau = 0$): (a) space/time plot of individual currents; (b) time series of a current difference.

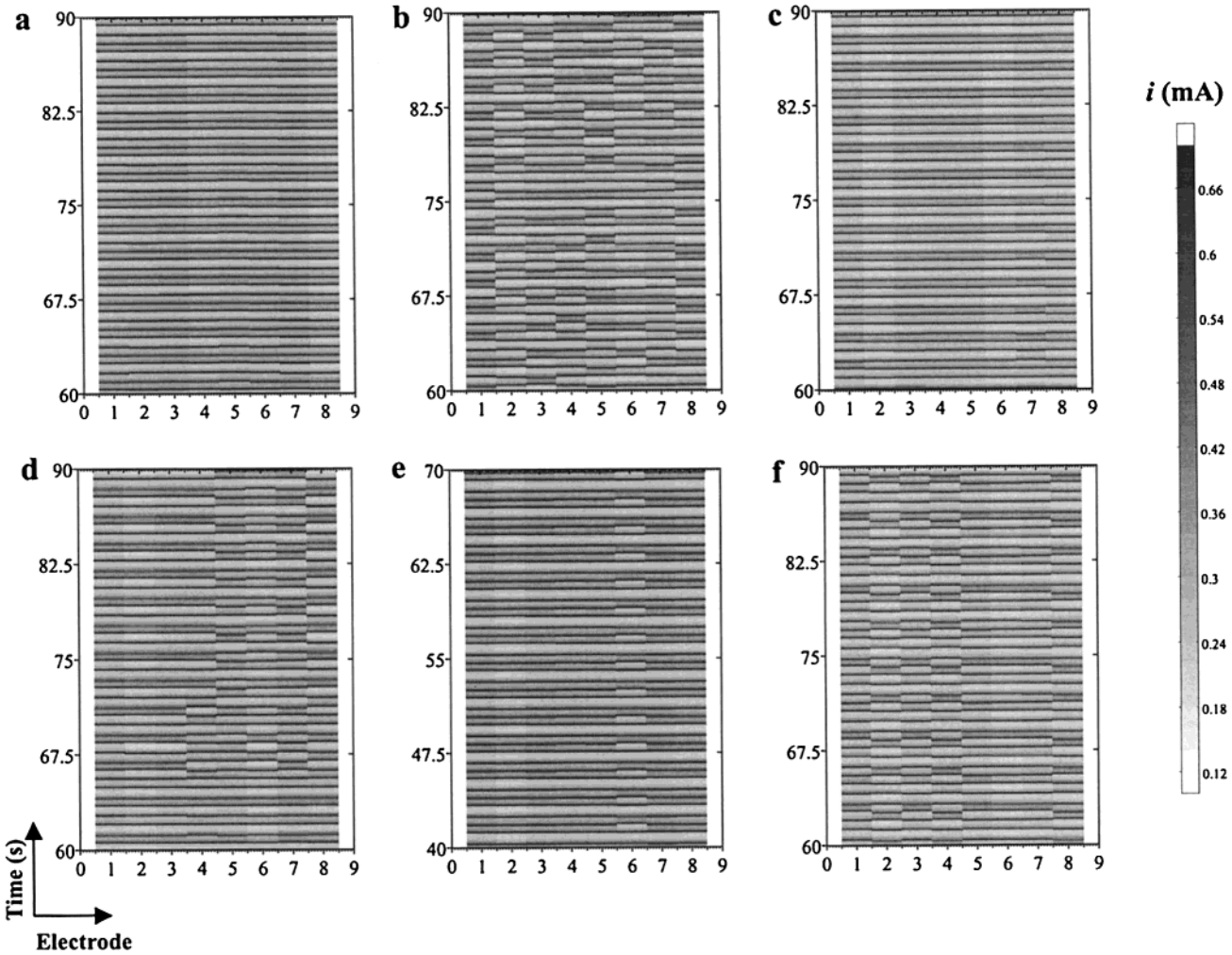

Figure 9. Space/time plot of individual currents: effects of delay time on an 8-electrode array ($\epsilon = 0.225$, $K = 12.5$ mV/mA, $R_{eq} = 906\ \Omega$, $V_0 = 1.355$ V): (a) synchronized period 3 state, $\tau = 0$ s; (b) $\tau = 0.1$ s; (c) synchronized period 3 state, $\tau = 0.75$ s; (d) intermittent clusters, $\tau = 0.8$ s; (e) stable period 3 clusters, $\tau = 1.55$ s; (f) stable period 6 clusters, $\tau = 2.35$ s.

three-dimensional state space is less than some value,[22] here taken to be 0.06 mA. The mean order parameter has a value of approximately zero for uncoupled chaotic oscillators and one in the synchronized state. We show this mean order parameter as a function of feedback gain in Figure 7. There is a wide range of feedback gain ($K = 1.8-3.6$ mV/mA) where stable chaotic and periodic clusters are formed. Note that there is a plateau at a value of the order parameter slightly above 0.5 in the plot. In this region the configuration of cluster sizes is balanced, viz., not too different from (32, 32). As the synchronized region is approached, one of the clusters grows and dominates the system. There is then a smooth monotonic increase of the order parameter to 1.0 at the synchronized state.

Transients. The behavior discussed above was all for the stationary state reached after transients had died down. The transient behavior before this stationary state does exhibit some general features, however, and is briefly discussed here. In all of the experiments, the feedback was turned on at $t = 20$ s. After the feedback is applied, the system has a tendency to synchronize, even at low feedback gains. The synchronization then either remains (high feedback gain) or breaks up (lower gain) to the previously described behaviors. We show a representative initial transient in the stable chaotic clustering region in Figure 8. As in all experiments, the feedback was turned on at $t = 20$ s and the elements quickly synchronize. However, after approximately 3 s, the synchronization breaks up into stable clusters. The current difference between electrodes 45 and 46 (Figure 8b) shows the transient. The initial state is unsynchronized chaos. The transient synchronization after $t = 20$ s causes a decrease in the difference in currents on the two electrodes. After approximately 2−3 s, the current difference increases as the synchronized state breaks up. Finally, however, the difference returns to near zero as the stable clusters are formed at $t = 27$ s.

Time Delay. We carried out some limited experiments on the effect of time delay using an 8-electrode array that consisted of one row of the larger array. In the 8-electrode array, behavior similar to that shown in Figure 2 for the 64-electrode array, that is, just below the clustering region, occurs at $\epsilon = 0.225$ (lower than that in the experiments with the 64-electrode array), and so we choose that value for these experiments.

The characteristic time scale of the chaotic oscillations against which the time delay is measured was obtained from a FFT power spectrum analysis of the individual chaotic signal without added global coupling. The spectrum has a main peak at about 1.33 Hz, which corresponds to a characteristic period of about 0.75 s.

A series of experiments carried out at fixed feedback gain ($K = 12$ mV/mA) and varying time delay is shown in Figure 9. This value of the feedback gain is large enough to synchronize the array at a zero time delay, as seen in Figure 9a; however, the periodic behavior is period 3 rather than the period 1 behavior seen above. A small time delay, such as 0.1 s is sufficient to break up the synchronized periodic state, as seen in Figure 9b. As the time delay is further increased to a value equal to the characteristic time of the chaotic oscillations, 0.75 s, another ordered state is obtained, as shown in Figure 9c where a synchronized periodic state is seen. At the delay value of 0.8 s (Figure 9d), which is somewhat higher than one characteristic period, the synchronization cannot be sustained and intermittent

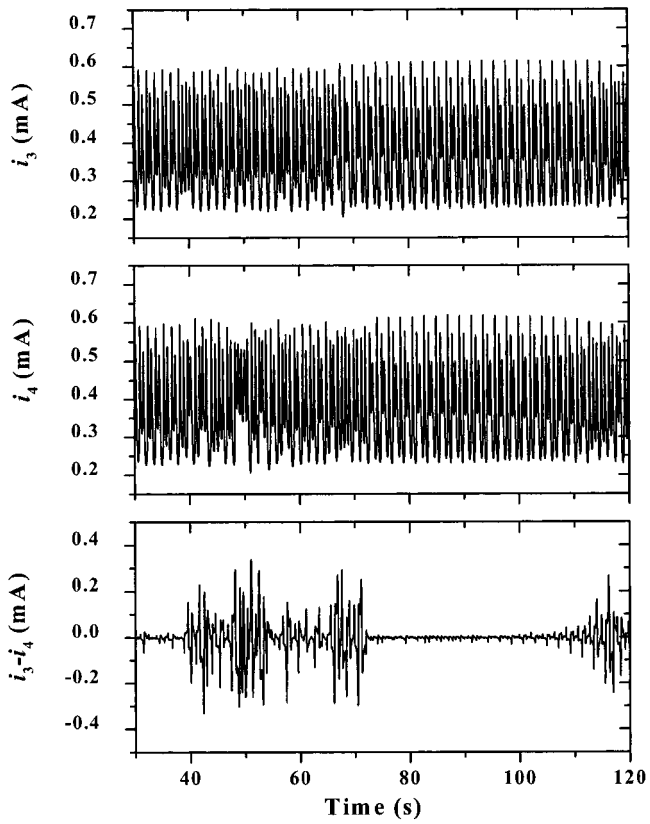

Figure 10. Time series of individual currents and differences between a pair showing intermittent clusters. The time series corresponds to Figure 9d (8-electrode array, $\tau = 0.8$ s, $\epsilon = 0.225$, $K = 12.5$ mV/mA, $R_{eq} = 906\ \Omega$, $V_0 = 1.335$ V).

clustering emerges. The current difference between two electrodes (Figure 10) shows this intermittency. The difference is very small at the time period between 34 and 38 s and again between 70 and 100 s, during which times the unstable clusters are formed. Three clusters are observed in a (5, 2, 1) configuration. At other time intervals shown in the figure, the elements diverge away from each other, resulting in a large current difference. We note that when the two electrodes are clustered, the individual time series of the two electrodes exhibit periodic oscillations (e.g., period 3 between 70 and 100 s). When the two electrodes diverge from each other, however, the individual time series are chaotic.

Ordered states are formed at time delays close to multiples of the characteristic period. Parts e and f of Figure 9 show period 3 clusters at a delay time of 1.55 s (about two characteristic periods) and period 6 clusters at delay time of 2.35 s (about three characteristic periods), respectively.

Concluding Remarks

A global coupling was imposed through feedback on a set of electrochemical oscillators. Without feedback the individual chaotic oscillators are weakly coupled. The gain of the feedback could be varied between zero, where the oscillators had almost independent dynamics, to a value at which the oscillators were synchronized. Thus, with increases in the feedback strength or gain, the system becomes more ordered; however, the transition to synchronization is not monotonic. As the feedback is increased from zero, intermittent, unstable clusters are first seen. This is followed by a state with stable chaotic clusters. We have observed only conditions with two

stable clusters, although, of course, many such configurations exist at the same parameter values.

With a further increase in the coupling strength past the stable chaotic cluster region, stable periodic clusters form. There are two-cluster states at lower gain, but three-cluster states arise as the gain is increased. As the gain becomes larger, one of the three clusters dominates and it occupies a larger and larger fraction of the reacting sites. Eventually, with higher gain, the one cluster takes over the entire region and the array is synchronized.

The feedback used here, in which a signal based on the total current is fed back to the applied potential, has two effects on the dynamics. First, it furnishes a global coupling. Second, the feedback influences the dynamics of the oscillators. There are thus some differences between the results of applying feedback to those obtained with the use of a global coupling not involving feedback.[37] With feedback, periodic states are observed. More than two clusters are obtained and, as noted above, the transition to the synchronized state is smooth, with one of the clusters dominating.

Both local (diffusive) and global interactions can affect the dynamics of nonlinear systems. The global interactions can be imposed through direct global coupling, through global feedback as in this study, and through globally imposed external forcing. In particular, the feedback with two control parameters—gain and delay time—provides a convenient way to change the system dynamics to the desired spatiotemporal behavior. A straightforward algorithm can be used in large systems with a relatively simple setup because only global information (the total current) and a global input (the imposed change in potential) are required. Although many theoretical studies have been carried out with examples in biology, physics, and chemistry, far fewer experimental studies are known. The electrochemical arrays used here thus represent a real chemical system in which the coupling can be carefully controlled and in which the spatiotemporal dynamics can be measured.

Acknowledgment

We thank John Quinn for the many years in which he has been an inspiring teacher, a supportive colleague, and, most importantly, a good friend. This work was supported by the National Science Foundation.

Literature Cited

(1) Kaneko, K. Spatiotemporal Chaos in one- and two-dimensional coupled map lattices. *Physica D* **1989**, *37*, 60.

(2) Carroll, T. L.; Heagy, J. F.; Pecora, L. M. Transforming signals with chaotic synchronization. *Phys. Rev. E* **1996**, *54*, 4676.

(3) Elson, R. C.; Selverston, A. I.; Huerta, R.; Rulkov, N. F.; Rabinovich, M. I.; Abarbanel, H. D. I. Synchronous behavior of two coupled biological neurons. *Phys. Rev. Lett.* **1998**, *81*, 5692.

(4) Rabinovich, M. I.; Abarbanel, H. D. I. The role of chaos in neural systems. *Neuroscience* **1998**, *87*, 5.

(5) Fei, Z.; Hudson, J. L. Pacemaker-Driven Spatiotemporal Patterns on an Electrode Array. *J. Phys. Chem.* **1997**, *101*, 10356.

(6) Ims, R. A.; Andreassen, H. P. Spatial Synchronization of Vole Population Dynamics by Predatory Birds. *Nature* **2000**, *408*, 194.

(7) Tuckwell, H. C.; Toubiana, L.; Vibert, J.-F. Spatial Epidemic Network Models with Viral Dynamics. *Phys. Rev. E* **1998**, *57*, 2163.

(8) Rulkov, N. F. Regularization of Synchronized Chaotic Bursts. *Phys. Rev. Lett.* **2001**, *86*, 183.

(9) Krischer, K. In *Modern Aspects of Electrochemistry*; Conway, B. E., Bockris, O. M., White, R. E., Eds.; Kluwer Academic/Plenum Press: New York, 1999; Vol. 32, p 1.

(10) Christoph, J.; Otterstedt, R. D.; Eiswirth, M.; Jaeger, N. I.; Hudson, J. L. Negative coupling during oscillatory pattern formation on a ring electrode. *J. Chem. Phys.* **1999**, *110*, 8614.

(11) Fei, Z.; Hudson, J. L. Chaotic Oscillations on Arrays of Iron Electrodes. *Ind. Eng. Chem. Res.* **1998**, *37*, 2172.

(12) Zeyer, K. P.; Holz, R.; Schneider, F. W. Continuous Coupling of Chaotic and Periodic States of Chemical Oscillators With and Without Time-Delay. *Ber. Bunsen-Ges. Phys. Chem.* **1993**, *97*, 1112.

(13) Dolnik, M.; Epstein, I. R. Coupled chaotic chemical oscillators. *Phys. Rev. E* **1996**, *54*, 3361.

(14) Huerta, R.; Bazhenov, M.; Rabinovich, M. I. Clusters of synchronization and bistability in lattices of chaotic neurons. *Europhys. Lett.* **1998**, *43*, 719.

(15) Zanette, D. H.; Mikhailov, A. S. Mutual synchronization in ensembles of globally coupled neural networks. *Phys. Rev. E* **1998**, *58*, 872.

(16) Ermentrout, B. Neural networks as spatio-temporal pattern-forming systems. *Rep. Prog. Phys.* **1998**, *61*, 353.

(17) Rabinovich, M. I.; Torres, J. J.; Varona, P.; Huerta, R.; Weidman, P. Origin of coherent structures in a discrete chaotic medium. *Phys. Rev. E* **1999**, *60*, R1130.

(18) Kaneko, K. Chaotic but regular Posi-Nega switch among coded attractors by cluster-size variation. *Phys. Rev. Lett.* **1989**, *63*, 219.

(19) Rosenblum, M. G.; Pikovsky, A. S.; Kurths, J. Phase Synchronization of Chaotic Oscillators. *Phys. Rev. Lett.* **1996**, *76*, 1804.

(20) Xie, F.; Cerdeira, H. A. Coherent-ordered transition in chaotic globally coupled maps. *Phys. Rev. E* **1996**, *54*, 3235.

(21) Just, W. On the collective motion in globally coupled chaotic systems. *Phys. Rep.* **1997**, *290*, 101.

(22) Zanette, D. H.; Mikhailov, A. S. Condensation in Globally Coupled Populations of Chaotic Dynamical Systems. *Phys. Rev. E* **1998**, *57*, 276.

(23) Vadivasova, T. E.; Balanov, A. G.; Sosnovtseva, O. V.; Postnov, D. E.; Mosekilde, E. Synchronization in driven chaotic systems: Diagnostics and bifurcations. *Phys. Lett. A* **1999**, *253*, 66.

(24) Middya, U.; Graham, M. D.; Luss, D.; Sheintuch, M. Pattern selection in controlled reaction−diffusion systems. *J. Chem. Phys.* **1993**, *98*, 2823.

(25) Steinbock, O.; Zykov, V.; Müller, S. C. Control of Spiral-wave Dynamics in Active Media by Periodic Modulation of Excitability. *Nature* **1993**, *366*, 322.

(26) Petrov, V.; Ouyang, Q.; Swinney, H. L. Resonant pattern formation in a chemical system. *Nature* **1997**, *388*, 655.

(27) Horváth, A. K.; Dolnik, M.; Munuzuri, A. P.; Zhabotinsky, A. M.; Epstein, I. R. Control of Turing structures by periodic illumination. *Phys. Rev. Lett.* **1999**, *83*, 2950.

(28) Vanag, V. K.; Yang, L.; Dolnik, M.; Zhabotinsky, A. M.; Epstein, I. R. Oscillatory cluster patterns in a homogeneous chemical system with global feedback. *Nature* **2000**, *406*, 389.

(29) Lev, O.; Wolffberg, A.; Sheintuch, M.; Pismen, L. M. Bifurcations to Periodic and Chaotic Motions in Anodic Nickel Dissolution. *Chem. Eng. Sci.* **1988**, *43*, 1339.

(30) Chang, M.; Schmitz, R. A. Feedback Control of Unstable States in a Laboratory Reactor. *Chem. Eng. Sci.* **1975**, *30*, 837.

(31) Chang, M.; Schmitz, R. A. An Experimental Study of Oscillatory States in a Stirred Reactor. *Chem. Eng. Sci.* **1975**, *30*, 21.

(32) Zimmermann, E. C.; Schell, M.; Ross, J. Stabilization of unstable states and oscillatory phenomena in an illuminated thermochemical system: Theory and experiment. *J. Chem. Phys.* **1984**, *81*, 1327.

(33) Kramer, J.; Ross, J. Stabilization of unstable states, relaxation, and critical slowing down in a bistable system. *J. Chem. Phys.* **1985**, *83*, 6234.

(34) Battogtokh, D.; Mikhailov, A. Controlling turbulence in the Complex Ginzburg−Landau Equation. *Physica D* **1996**, *90*, 84.

(35) Parmananda, P.; Eiswirth, M. Suppression of Chemical Turbulence Using Feebacks and Forcing. *J. Phys. Chem. A* **1999**, *103*, 5510.

(36) Kiss, I. Z.; Wang, W.; Hudson, J. L. Experiments on arrays of globally coupled periodic electrochemical oscillators. *J. Phys. Chem. B* **1999**, *103*, 11433.

(37) Wang, W.; Kiss, I. Z.; Hudson, J. L. Experiments on arrays of globally coupled chaotic electrochemical oscillators: Synchronization and clustering. *Chaos* **2000**, *10*, 248.

Received for review January 25, 2001
Accepted March 6, 2001

IE0100737