Supporting Online Material for

Engineering Complex Dynamical Structures: Sequential Patterns and Desynchronization

István Z. Kiss, Craig Rusin, Hiroshi Kori, John L. Hudson

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Supporting Online Material

A) Design of the feedback: analytical results

1) The model
We consider a population of $N$ oscillators under a global feedback, reading

$$\frac{dA_i(t)}{dt} = F[A_i(t)] + KP(t),$$  \hspace{1cm} (1)

where $A_i = (x_i, y_i, \ldots)\text{T}$ is the state variable of the oscillator $i$ ($i = 1, 2, \ldots, N$), $F(\cdot)$ is a non-linear function describing a limit-cycle oscillation, $P(t)$ is the non-linear function expressing the non-linear, time-delayed global feedback. We assume that $P(t)$ is constructed by and applied to a certain element of the state variables. In particular, we introduce the following feedback:

$$P(t) = \left( \frac{1}{N} \sum_{j=1}^{N} p_j(t), 0, 0, \ldots \right)^\text{T},$$  \hspace{1cm} (2)

$$p_j(t) = \sum_{n=0}^{\infty} k_n \left[ x_j(t - \tau_n) - a_0 \right]^n,$$  \hspace{1cm} (3)

where $k_n$ and $\tau_n$ are respectively the intensity and the time-delay of $n$-th polynomial feedback, and $a_0$ is the mean value of $x_j$ of the uncoupled system (i.e., $K = 0$). Our objective of this supplementary material is to provide a recipe for designing the interaction function introduced below, which characterizes the dynamical behavior of the oscillator population, via our control parameters $k_n$ and $\tau_n$.

2) General formula
We first define the phase $\phi$ of oscillation. We consider the unperturbed system

$$\frac{dA}{dt} = F(A)$$  \hspace{1cm} (4)

By assumption, the system asymptotically converges to a stable periodic (i.e., limit-cycle) solution $A^0(t)$ for $t \rightarrow \infty$. Its intrinsic period is defined as $T$ (and the intrinsic frequency is $\omega = 2\pi/T$), so that $A^0(t + T) = A^0(t)$. The phase $\phi(A)$ of the state $A$ can be defined not only on the limit-cycle but on the global state space in such a way that the following condition is satisfied:

$$\frac{d\phi(A)}{dt} = \omega,$$  \hspace{1cm} (5)

To complete the definition, we have to give a value of $\phi$ at some state point $A$. This can arbitrarily be chosen to our convenience.

Under the weak coupling condition (i.e., for small $K$), we may apply the phase reduction method (1) and reduce the system (1) to the standard form of the coupled phase oscillator system in the first order approximation for $K$. We then obtain

$$\frac{d\phi(t)}{dt} = \omega + \frac{K}{N} \sum_{j=1}^{N} H[\phi_j(t) - \phi(t)],$$  \hspace{1cm} (6)

Here, the $2\pi$-periodic function $H(\cdot)$, called the interaction function, is defined as
\[ H(\phi_j - \phi_i) = \frac{1}{2\pi} \int_0^{2\pi} Z(\phi_j + \lambda) p(\phi_j + \lambda) d\lambda = \frac{1}{2\pi} \int_0^{2\pi} Z(\lambda) p(\phi_j - \phi_i + \lambda) d\lambda, \]  

(7)

where \( Z(\cdot) \) is the \( 2\pi \)-periodic function called the phase response function, and \( p(\phi_j) \) is the feedback signal \( p_j(t) \) from the oscillator \( j \) described in terms of the phase \( \phi_j \). The functions \( Z(\cdot) \) and \( p(\cdot) \) are obtained from the unperturbed system (4) in the following manner. The response function \( Z(\phi) \) is the gradient of the phase along the \( x \)-direction on the limit-cycle orbit, i.e.,

\[ Z(\phi) = \frac{\partial \phi}{\partial x} \bigg|_{A-e}. \]  

(8)

For convenience, we expand

\[ Z(\phi) = \sum_l z_l e^{il\phi}. \]  

(9)

To compute \( p(\phi) \), we also expand the \( 2\pi \)-periodic trajectory \( x(\phi) \) on the limit-cycle orbit \( A^0 \) as

\[ x(\phi) = \sum_l a_l e^{il\phi}. \]  

(10)

Note that \( \zeta_l = \zeta_{-l} \) and \( a_l = a_{-l} \). Furthermore, because \( \phi(t - \tau) = \phi(t) - \omega \tau \), we obtain

\[ x(\phi(t - \tau)) = \sum_l a_l e^{il\phi(t)} e^{-il\omega \tau}. \]  

(11)

Thus, the feedback signal \( p(\phi) \) is described as

\[ p(\phi) = \sum_n k_n \left\{ x(t - \tau_n) - a_0 \right\} = \sum_n k_n \left\{ \sum_l a_l e^{il\phi} e^{-il\omega \tau} \right\}^n. \]  

(12)

Hence, we may design a desired interaction function \( H_{\text{large}}(\phi) \) by taking the following procedure. (i) Measurement of \( Z(\phi) \) and \( x(\phi) \). The function \( Z(\phi) \) can be found numerically (or in some cases analytically) from dynamical equations or experimentally. For example, see Refs. (1-3) for the analytical derivation of \( Z(\phi) \), Refs. (4, 5) for the numerical derivation, and Refs. (6, 7) for the experimental derivation. (ii) Fourier expansions of \( Z(\phi) \) and \( x(\phi) \) up to certain harmonics and calculation of \( H(\phi) \) through Eq. (7). (iii) Determination of \( k_n \) and \( \tau_n \) by comparison of \( H(\phi) \) with \( H_{\text{large}}(\phi) \). An illustrative example is provided in the next section.

3) Oscillators with nearly harmonic signals

We consider limit-cycle oscillators generating nearly harmonic signal \( x(\phi) \), i.e., \( |a_i| \geq |a_l| \) for \( l \geq 2 \). In this case, we may obtain a simpler expression of the interaction function \( H(\phi) \).

By the assumption \( |a_i| \geq |a_l| \) for \( l \geq 2 \), we may approximate

\[ \{ x(\phi) - \bar{x} \}^n \approx (a_l e^{i\phi} + a_{-l} e^{-i\phi})^n = \sum_{m=0}^n C_m a_{l}^m a_{-l}^{n-m} e^{i(2m-n)\phi}, \]  

(13)

which is followed by

\[ p(\phi) = \sum_n k_n \sum_{m=0}^n C_m a_{l}^m a_{-l}^{n-m} e^{i(2m-n)(\phi - \omega \tau_n)}. \]  

(14)

\((C_m \text{ is a number of } m \text{ combination from a set of } n \text{ elements})\). Then, from Eq. (7), we get
\[ H(\phi) = \sum_{n} k_n \sum_{m=0}^{n} C_n \alpha_n m a_1^{m} a_{-1}^{n-m} e^{i(2m-n)k\phi + \text{c.c.}}. \] (15)

The \( n \)-th polynomial feedback thus enhances \( l \)-th harmonics (where \( l = n, n-2, n-4, \ldots \)). Therefore, if a target interaction function consists of up to \( n \)-th harmonics, we need up to \( n \)-th polynomials in the feedback signal (3).

As an illustration, suppose that our target function is of the following form

\[ H_{\text{target}}(\phi) = \sin(\phi - \alpha) - r \sin(2\phi) = \frac{1}{2} e^{-i(\alpha + \frac{\pi}{2})} e^{i\phi} + \frac{r}{2} e^{i\phi} e^{-i\phi} + \text{c.c.}, \] (16)

(where \( \alpha \) and \( r \) are the parameters of the function). For this target function, we need up to quadratic signal, where Eq. (15) becomes

\[ H(\phi) = k_0 + 2k_2 |a_1|^2 z_0 + k_1 e^{-i\omega r} a_1 z_{-1} e^{i\phi} + k_2 e^{-i2\omega r} a_1^2 z_{-2} e^{i2\phi} + \text{c.c.}. \] (17)

Comparing Eq. (16) and Eq. (17), we find

\[ k_0 = -z_0 \left| \frac{r}{z_{-2}} \right|, \quad k_1 = \frac{1}{2a_1 z_{-1}}, \quad k_2 = \frac{r}{2a_1^2 z_{-2}}, \] (18)

\[ \tau_1 = \alpha + \frac{\pi}{2} + \text{arg}(a_1 z_{-1}) \omega \frac{1}{|a_1 z_{-2}|}, \quad \tau_2 = -\frac{\pi}{2} + \text{arg}(a_1^2 z_{-2}) - \text{arg}(r) \frac{1}{2 \omega}. \] (19)

Note that we can also use the sets \((-k_1, \tau_1 + \pi/\omega)\) instead of \((k_1, \tau_1)\) and/or \((-k_0, -k_2, \tau_2 + \pi/2\omega)\) instead of \((k_0, k_2, \tau_2)\).
B) Measurement of the Response Function

The response function for the electrochemical oscillators was determined using the equation:

\[
H(\Delta \phi) = \frac{1}{2\pi} \int_{0}^{2\pi} Z(\phi) h(x(\phi + \Delta \phi)) \, d\phi
\]  

(20)

where \( H \) is the interaction function, \( Z \) is the response function, \( h \) is the coupling function (i.e., the feedback signal) and \( x \) is the waveform. For the purpose of this paper, the coupling function is a time delayed polynomial feedback function of the form

\[
h(x) = \sum_{n=0}^{S} k_n x(t - \tau_n)^n
\]

(21)

where \( k_n \) is the \( n^{th} \) polynomial coefficient, \( x \) is the zero-mean waveform which is scaled by its amplitude, \( \tau_n \) is the time delay, and \( S \) is the polynomial feedback order. These equations allow the response function to be calculated from a measured interaction function under known feedback conditions.

Experimental Measurement of an Interaction Function

Interaction functions were experimentally measured using the method developed by J. Miyazaki and S. Kinoshita (8). In this method, the frequency of 2 slightly dissimilar oscillators was recorded as they process around each other over time. The interaction function was obtained by calculating the change in frequency of each oscillator as a function of the phase difference between the two elements. It is important to note that the magnitude of the interaction between oscillators must be strong enough such that the frequency change can be accurately measured, but weak enough to avoid causing the elements to synchronize.

For example, to produce the response functions found in Fig. 1C of the manuscript, a population of 64 oscillators was created at 1.165V and allowed to line out for a period of 2 hours. Two elements in the population were selected such that their frequency difference was \(~3\%\). A feedback signal was then applied to the system, with the parameters: \( k_0 = -0.01 \text{V}, k_1 = 1.366, \tau_1 = 0.21 \text{rad}/2\pi, k_2 = 3.899 \text{V}^{-1}, \tau_2 = 0.61 \text{rad}/2\pi, K = 0.0005 \). The numerical values of the feedback parameters were chosen such that 1) they did not disturb the shape of the waveform, 2) they did not cause synchronization, and 3) they produced a

![Fig S1: Measuring an interaction function. A) Unbounded phase difference of 2 heterogeneous smooth oscillators (1.165 V). B) Fluctuation of the oscillator period over time due to a nonlinear feedback. \( k_0 = -0.01 \text{V}, k_1 = 1.366, \tau_1 = 0.21 \text{rad}/2\pi, k_2 = 3.899 \text{V}^{-1}, \tau_2 = 0.61 \text{rad}/2\pi, K = 0.0005 \). C) Zero mean interaction function obtained from the data in panels A and B using equation 22.](image)
measurable change in the oscillator period. Any feedback parameters that obey the constraints could be selected, since the response function remains constant under all viable feedback conditions.

![Image](image_url)

**Fig. S2:** Measured interaction function data which were used to regress the response function for smooth and relaxational oscillators. A) Interaction function for smooth oscillator (1.165V) with feedback parameters: $k_0=-0.01V$, $k_1=1.366$, $k_2=3.899 V^{-1}$, $\tau_1=0.21 rad/2\pi$, $\tau_2=0.61 rad/2\pi$ B) Interaction function for smooth oscillator (1.165V) with feedback parameters: $k_0=-0.24V$, $k_1=8.4547$, $k_2=20.3496 V^{-1}$, $\tau_1=0.12 rad/2\pi$, $\tau_2=0.44 rad/2\pi$ C) Interaction function for a relaxational oscillator (1.225V) with feedback parameters: $k_0=-0.1$, $k_2=-1 V^2$, $\tau_3=0.01 rad/2\pi$. The lines represent the how well the optimized response functions reproduced the original measured interaction functions.

The feedback signal caused the period of the oscillators to fluctuate as the phase difference between the two elements changed. This effect is illustrated in figure S1. After collecting approximately 200 cycles of data, the interaction function was calculated using the equation:

$$H(\Delta\phi) = \frac{-2\pi}{EP_{base}^2} \left[ P(\Delta\phi) - P_{base} \right]$$

where $E$ is the overall gain of the feedback, $P_{base}$ is the inherent frequency of a single oscillator (in this case 2.04s), and $P(\Delta\phi)$ represents the period of the oscillator as a function of the phase difference between the two elements. The function $P(\Delta\phi)$ was easily found by measuring the time period between adjacent waveform peaks. The phases of the oscillators were reconstructed based on peak to peak interpolation on the interval $[0, 2\pi]$. The analysis assumes that the phase difference between the elements does not change over one oscillation cycle, which was an acceptable approximation since the inherent frequency difference was only $\sim$3%.

**Response Function Calculation**

Continuing the above example of Figure 1C, two interaction functions were measured for the experimental system at 1.165V, under different feedback conditions. These sets are illustrated in Fig. S2, panels A and B. These sets were used in a multi-objective optimization to find the single best response function which can reproduce both sets of interaction functions simultaneously. For the purpose of this paper, the Fourier coefficients of the response function were used as the optimization parameters.

The number of coefficients to be optimized was determined by the number of higher harmonics present in the waveform and the measured interaction functions. When the system was close to the Hopf bifurcation point (~1.105V) only 1-2 coefficients were necessary, as the response function was nearly sinusoidal. However, when the voltage was increased, the system became relaxational (~1.20V), and the response function required $\sim$5-7 coefficients. In the case of figure 1C, the number of Fourier coefficients was set to 3, since the system exhibited smooth oscillations, but was at a voltage (1.165V) higher than the
Hopf bifurcation point. A simplex optimization algorithm was used to determine their value by minimizing an objective function:

\[
\text{error} = \left[ \sum_{n=1}^{N} \sum_{i=1}^{Pts} \left( T_{i}^{n} - D_{i}^{n} \right) \right]^{\frac{1}{2}}
\]

(23)

where \( T_{i}^{n} \) is the \( i \)th data point in the \( n \)th measured interaction function data set, \( D \) is the optimized interaction function based on the optimized Fourier coefficients of the response function, \( N \) is the number of measured interaction function data sets, and \( Pts \) is the number of measured points in the function. The initial conditions were taken to the Fourier coefficients of a sin wave, since the response function becomes a sin wave as the system approaches the Hopf bifurcation point. Thus, the algorithm generated a single response function which was capable of reproducing all of the measured interaction functions, which is shown in figure 1C. This simultaneous fitting was necessary to eliminate the effects of experimental noise in the measured interaction functions.

Alternatively, the Fourier coefficients of the response function can be directly obtained via equation 20 by expanding the response function \( Z \), the feedback function \( h \), and the interaction function \( H \), in Fourier series. This was the method used to determine the response function for the relaxational oscillator, using the interaction function in Fig S2 panel C. Integrating through, and solving for the Fourier coefficients of the response function we obtain:

\[
Z_{k}^{*} = H_{k} / h_{k}
\]

(24)

where \( Z_{k}^{*} \) is the complex conjugate of the Fourier coefficients of the response function, \( H_{k} \) are the Fourier coefficients of the interaction function (experimentally measured), and \( h_{k} \) are the Fourier coefficients of the feedback signal, calculated using equation 21.

C) Design of Interaction Functions

Interaction Function for Phase Locked Oscillators

Consider an interaction function with the following odd component:

\[ H(\Delta \phi) = \sin(\Delta \phi) + R \sin(2 \Delta \phi) \]

In two identical oscillators the phase locked solutions \((\Delta \phi^{*})\) from Eq. 3 of the manuscript are:

I. trivial solutions: 0 and \( \pi \)

II. non-trivial solutions satisfy: \( R = -\sin(\Delta \phi^{*}) / \sin(2\Delta \phi^{*}) \)

(These states can be made stable by the proper sign of the overall feedback gain \( K \).) In the manuscript, we took a special case of \( \Delta \phi^{*} = \pi / 2 \). This results in \( R \to -\infty \) corresponding to an odd component that has purely second harmonics

\[ H(\Delta \phi) = -\sin(2 \Delta \phi) \]

This is only the odd component of the interaction function. As far the even component is concerned, we added the simplest, first harmonic cosine term. We found that such an addition greatly improves the robustness of the fitting procedure.

Interaction Function for Slow Switching

Consider an interaction function with the following structure

\[ H(\Delta \phi) = \sin(\Delta \phi + \alpha) - R \sin(2 \Delta \phi) \]
Setting the values of the parameters to $\alpha = -1.25$ and $R = 0.25$ yields Hansel’s function which has been shown to admit heteroclinic connections between unstable fixed points. With 4 oscillators, these cluster states are separated by a phase difference of $\sim 0.18 \text{ rad}/2\pi$. Our goal was to obtain a parameter set which would:

I. Increase experimental robustness against heterogeneity (9)
II. Increase the phase difference between the unstable cluster states such that two distinct cluster states could be easily identified.

The final parameter set: $\alpha = -1.32$ and $R = 0.25$ is a balance between achieving the two stated objectives and maintaining the heteroclinic connections. Our phase modeling indicated that the phase difference between cluster states increased from 0.18 to 0.22 rad/2$\pi$, (a 22% increase), while the slow switching dynamics remained intact. When $\alpha$ was set above this point, slow switching was difficult to observe experimentally.

**Optimal Interaction Function for Desynchronization**

We wish to design an interaction function with negative sin terms in order to destabilize cluster states. We observed both two- and three-cluster states with linear feedback. (Only the three-cluster state is mentioned in the main text.) The existence of the two- and three-cluster states induced by the linear feedback implies the importance of second and third harmonics in $H$ which can be manipulated by quadratic and cubic feedbacks. Therefore, we sought an interaction function with feedback parameters $k_1$, $\tau_1$, $k_2$, $\tau_2$, $k_3$, $\tau_3$ in such a way that

- The odd part of the interaction function is composed of negative sin terms up to five harmonics. Negative fourth and fifth order harmonics are utilized to avoid inadvertent four- and five-cluster states. (Harmonics higher than five are weak and do not induce clusters in our system.)
- The feedback produces a weak perturbation signal $\delta p(t)$ (Eqs. 1 and 2 in the text) measured by its power.

The optimization requires adjustment of 6 parameters. A two-step procedure was applied:

I. Obtaining the feedback delays:
   Since the linear feedback with small delay destabilized the one-cluster state, we set the linear delay to $\tau_1 = 0.01 \text{ rad}/2\pi$. (Smaller values are difficult to implement in the experimental system because of real-time filtering.)

   For the delay of the second order feedback the interaction function was determined for a purely quadratic feedback with $k_2 = 1 \text{ V}^{-1}$ at various values of $0.01 < \tau_2 < 0.12 \text{ rad}/2\pi$ (The phase model approximation for large delays may fail.) A value $\tau_2 = 0.09 \text{ rad}/2\pi$ was found where the sum of the sign of the first five harmonics of $H$ with the quadratic feedback has a maximum (corresponding to positive feedback) or minimum (negative feedback).

   The delay of the third order feedback was determined similarly to that of the second order but using purely third order feedback in the calculation: $\tau_3 = 0.09 \text{ rad}/2\pi$.

II. Obtaining the feedback gain
   With equal values of the gains ($k_1$, $k_2$, $k_3$) a purely linear feedback was approximately 10 times stronger than a quadratic, measured by the power of the feedback signal; similarly, the quadratic was approximately ten times stronger than the cubic. Therefore, in order to obtain a mild feedback that desynchronizes the system, we minimized the cost function:

$$100k_1 + 10k_2 + k_3$$

with conditions that the odd part of the overall interaction function ($H = \sum b_k \sin(k\Delta\phi)$) satisfies

- $b_1 < -0.4$
- $b_2 < -0.2$
- $b_3 < -0.2$
- $b_4 < -0.05$
In order to destroy the synchronized (cluster-1) state, \( b_1 \) is chosen as the largest value. Linear feedback induces two- and three-cluster states so that \( b_2 \) and \( b_3 \) are somewhat smaller. The coefficients \( b_4 \) and \( b_5 \) have small negative values in order to avoid inadvertent four- and five-cluster states. The above defined optimization problem was solved with linear programming with the following result: \( k_1=1.09, k_2=-5.35 \text{ V}^{-1}, k_3=0 \text{ V}^{-2} \). The iterations to obtain the feedback parameters with linear programming converge very fast (it took less than one second on a desktop PC). Convergence is not significantly affected by heterogeneities between oscillators or noise in the system; however, these effects do play a role in the selection of proper set of target interaction functions. We chose a low-power feedback signal (see the cost function) optimized for desynchronization that can effectively suppress inherent noise and heterogeneities in the system.

D) Calculation of Feedback Parameters

Once the response function, waveform, and target interaction function are known, the feedback parameters which give rise to a particular interaction function can be calculated from equation (20). Two approaches have been developed to determine feedback parameters, a direct nonlinear optimization, and a linear programming algorithm. The nonlinear optimization method was used to determine feedback parameters for phase locking and slow switching experiments, while the linear programming algorithm was used in the desynchronization work. Since the linear programming algorithm was discussed in detail in the context of determining a desynchronizing interaction function (section C), it will not be discussed here.

The direct nonlinear optimization method minimized the error between the calculated and desired interaction functions by manipulating the feedback parameters (\( k_n \) and \( \tau_n \)) using a simplex algorithm. The objective function was

\[
error = \left[ \frac{1}{P_{\text{ts}}} \sum_{i=1}^{P_{\text{ts}}} \left| T_i - H_i \right| \right]^\frac{1}{2}
\]

(25)

where \( T_i \) is the \( i^{\text{th}} \) data point of the target interaction function, \( H \) is the interaction function calculated from the optimization parameters (i.e. the feedback parameters) via equations 20 and 21, and \( P_{\text{ts}} \) is the number of data points in the functions \( T \) and \( H \).

A pre-set number of feedback parameters (i.e. the feedback order) must be selected before the optimization can begin. The order of the feedback signal was set to the highest harmonic present in either the response function or the interaction function. This insures that the feedback signal was robust enough to cancel out any unwanted higher harmonics present in the system. The detailed mathematics of this fact are discussed in section A. Therefore, it was determined that second order feedback was necessary for the phase locking and slow switching experiments because of the second order component in the interaction function. The initial conditions for the optimization were taken to be \( k_n=10^{n-1} \) and \( \tau_n=0.5 \text{ rad/2\pi} \). This allows each polynomial feedback term to be the same order of magnitude, since \(|x|<1\). For slow switching, this produced the feedback parameters: \( k_1=8.7376, k_2=16.3696 \text{ V}^{-1}, \tau_1=0.21 \text{ rad/2\pi}, \tau_2=0.68 \text{ rad/2\pi} \) which were then used in the experimental system. The convergence of the fitting algorithm is very fast (in this case it took few seconds on a desktop PC). The convergence is not significantly affected by heterogeneities between oscillators or noise in the system; we picked an overall interaction for the slow-switching experiment that does not exhibit extreme sensitivities (9) to heterogeneities (see Section C).

E) Experimental Setup and Methodology
The experimental apparatus consisted of an electrochemical cell connected in series to a set of zero resistance ammeters (ZRA), which were attached to a real time data acquisition system. The cell was made up of 64 Ni electrodes (99.99% pure) in a 3M H₂SO₄ solution, a Pt mesh counter electrode, and a Hg/Hg₂SO₄/K₂SO₄ (sat) reference electrode. The cell was enclosed in a jacketed glass vessel, which allowed cooling water to surround the system, maintaining a constant temperature of 11°C. A diagram of the apparatus can be found in figure 1. A EG&G Princeton Applied Research Potentiostat was used to adjust the circuit potential (V) of the cell, causing the Nickel electrodes to undergo transpassive dissolution. The dissolution current of each electrode was measured with 64 ZRAs. In the absence of coupling, a 650Ω resistor was attached to each channel to induce oscillations. When coupling was required, a single coupling resistor (Rₗ) was attached in series to the common ground point of the ZRA, while the parallel resistors (Rₚ) were adjusted such that the overall resistance of the cell remained constant. The ratio between the parallel and series resistors determines the magnitude of the global coupling. (10) Without the series resistor, the oscillators experience no global coupling and are directly influenced by the feedback signal being applied to the system. A Labview based real time data acquisition computer was used to read the ZRA measurements, stream these measurements to the host machine, calculate the feedback signal, and apply it to the potentiostat at 250Hz. The feedback signal was calculated from the current measurements by first determining the electrode potential using the equation

\[ I(t) = \frac{V(t) - E(t)}{R} \]  

(S1)

where \( I(t) \) is the measured current, \( V(t) \) is the applied voltage, \( E(t) \) is the electrode potential, and \( R \) is the channel resistance. After finding the electrode potential, each element had its mean value removed and was scaled by its amplitude to eliminate any bias. The host machine was used to continuously determine offset and amplitude for each element in the population.

Before the Ni electrode is placed into the system, it was polished with a wet grinder to remove any initial oxide layer that may be present. Six polishing disks were used, decreasing in roughness from 180 grit to 4000 grit. This also ensured that the system started from approximately identical initial conditions for every experiment. After polishing, the electrodes were placed in the acid solution, and the potential was ramped from \(-0.68\)V to \(1.25\)V and back to \(0\)V without any resistors present. This was done to form a thin passive layer on each electrode. After this, all of the necessary resistors were connected and the system was brought up to the desired operating voltage. The system was allowed to line out at this voltage for \(~1.5\)–\(~2.0\) hours, as this was found to reduced the amount of frequency drift experienced by the oscillators.

Fig. S3: Experimental Setup.
Supporting Figure

**Figure S4:** Engineering a system of four oscillators to generate sequential cluster patterns: inter-cluster transition. Time series of the order parameter, \( R_k = \exp(ik\phi) \) (bottom panel), along with two corresponding cluster configurations (top panels). Feedback parameters \( k_0=-0.0526V, k_1=8.7376, k_2=16.3696 \ V^{-1}, \tau_1=0.21, \tau_2=0.68, K=0.0494, (V=1.165V, R_{tot} = 162.5 \ \Omega) \). Cluster elements are labeled for clarity.
Supporting References