Reconstruction of two-dimensional phase dynamics from experiments on coupled oscillators

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Phase models are a powerful method to quantify the coupled dynamics of nonlinear oscillators from measured data. We use two phase modeling methods to quantify the dynamics of pairs of coupled electrochemical oscillators, based on the phases of the two oscillators independently and the phase difference, respectively. We discuss the benefits of the two-dimensional approach relative to the one-dimensional approach using phase difference. We quantify the dependence of the coupling functions on the coupling magnitude and coupling time delay. We show differences in synchronization predictions of the two models using a toy model. We show that the two-dimensional approach reveals behavior not detected by the one-dimensional model in a driven experimental oscillator. This approach is broadly applicable to quantify interactions between nonlinear oscillators, especially where intrinsic oscillator sensitivity and coupling evolve with time.

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I. INTRODUCTION

Systems of coupled oscillators have been investigated in a variety of fields. Examples include coupled lasers [1], population dynamics [2], chemical reactions [3], and cardiorespiratory interactions [4,5], among others. A theoretical description of the system can be obtained in two ways: either write the model equations for the coupled systems starting from the first principles, or reconstruct the model equations from observations. In many cases, e.g., in biological systems, use of the former approach is greatly impeded by underlying complexity and lack of knowledge about oscillation generation and coupling mechanisms.

In this paper we follow the second approach and reconstruct the interaction between a pair of experimental nonlinear electrochemical oscillators. We discuss the basic theory, which we apply for our system of two oscillators with weak coupling. The system is represented in terms of two phases, which in many cases may be simplified to a single variable, the phase difference [6]. We show that phase models that preserve dependence on individual phases generally provide a more detailed description of the interactions between two oscillators than do those based on the phase difference. We compare results of the two modeling methods and discuss limitations of models based on phase difference. We calculate from experimental data a two-phase model using a previously introduced technique [7]. Our results experimentally verify phase reconstruction in a system with noise and connect the two-dimensional and one-dimensional models [6,8,9]. We calculate the natural oscillator frequencies, changes in coupling directionality, and coupling time delay from the experimentally determined phase models. We also present experiments where coupling functions exhibit higher-order terms, and show that these terms are not captured by the one-dimensional model.

II. THEORY

Suppose we observe two interacting oscillators described by

\[ \dot{x}_i = F_i(x_i) + \varepsilon \tilde{p}_i(x_i, \bar{x}_j), \]

where \( i = 1,2, \ j = 2,1, \) and the parameter \( \varepsilon \) describes the strength of the interaction. Generally, the functions \( F_i \) are different; moreover they can be of different dimension. The coupling functions \( \tilde{p}_i \) can be different as well. We assume that both systems when uncoupled, i.e., when \( \varepsilon = 0 \), possess stable limit cycles in their phase spaces. The asymptotic dynamics of each oscillator (after transients die out) can be then described by a single variable, the phase [6,9,10].

Even when the equations (1) of the coupled oscillating system are known their analytical treatment can be quite complicated, if at all possible. An essential simplification can be made in the case of weak coupling, where applied perturbations are small compared to the negative Lyapunov exponent(s) of each oscillator. For this case the oscillators remain near their closed orbits and the dynamics of a pair of coupled systems is confined to the two-dimensional torus in the phase space. Correspondingly, the dynamics can be parametrized by two phases [6,9,10],

\[ \dot{\phi}_i = \omega_i + Q^{(i)}(\phi_i, \phi_j). \] (2)

Here \( \phi_i \) is the phase of oscillator \( i, \phi_j \) is the phase of the other oscillator, and \( \omega_i \) is the natural angular frequency of the oscillator \( i \), i.e., the frequency of the uncoupled system. The functions \( Q^{(i)} \) describe the coupling between the systems. The only \( a \ priori \) assumption about these functions is that they are \( 2\pi \) periodic with respect to both arguments; in particular, they can contain a constant term.

If the dynamical equations (1) are known, the coupling functions \( Q^{(i)} \) can be represented in the form of the power series by means of a perturbative expansion [6],

\[ \dot{Q}^{(i)}(\phi_i, \phi_j) = \varepsilon Q_1^{(i)}(\phi_i, \phi_j) + \varepsilon^2 Q_2^{(i)}(\phi_i, \phi_j) + \cdots, \] (3)

where the subscripts on \( Q^{(i)} \) correspond to the order of approximation. Computation of the high-order terms represents, to the best of our knowledge, an unsolved problem, whereas the first-order phase approximation is widely used in various applications [6,8]. The first-order coupling functions can be written as

\[ Q_1^{(i)}(\phi_i, \phi_j) = \tilde{Z}_i(\phi_i) \cdot \tilde{h}_i(\phi_i, \phi_j), \] (4)
where $\tilde{Z}$ is the phase-dependent response function of the oscillator and $h = \mathcal{P}(\tilde{Z}(\phi_i, \tilde{X}_j(\phi_j)))$ is the applied stimulation. In the simplest case when the scalar driving is independent of the phase of the driven system and enters the state-space equations (1) as an additive term, the coupling function can be represented as a product of two functions of one variable, $Q_i^{(j)}(\phi_i, \phi_j) = Z_i(\phi_i) h_i(\phi_j)$. The phase description Eq. (2) can be valid for not-so-weak coupling as well: as long as a stable invariant torus in the phase space exists, the motion on it can be parametrized by the phases and the dynamics can be written in the form of Eq. (2).

A large body of work concerns the description of two interacting oscillators as a function of the phase difference [11]. Theoretical studies and numerical simulations show that these one-dimensional phase models capture the important synchronization properties of populations of similar oscillators with weak interactions [12–18]. Phase difference based phase synchronization properties of populations of similar oscillators of the phase of the driven system and enters the state-space where

\[
Q_{m,n}(\phi_i, \phi_j) = \mathcal{P}(\tilde{Z}(\phi_i)) h_i(\phi_j). \tag{7}
\]

Thus, a description in terms of phase difference is possible only in vicinities of the resonant frequency ratios. For each of the resonant tongues one should establish an averaged coupling function $q_{m,n}$. Thus, although a complete description of the coupled system for any frequency ratio can be achieved with one pair of two-dimensional functions $Q$, a large set of one-dimensional coupling functions is required to provide the same result.

We illustrate the difference in synchronization predictions between two- and one-dimensional phase models by an analysis of the following toy model of a harmonically driven oscillator:

\[
\begin{align*}
\dot{\phi}_1 &= \omega_1 + \varepsilon \left[ \cos(\phi_1) + \cos(2\phi_1) \right] \sin(\phi_2), \tag{7} \\
2\varepsilon x_1 \dot{\phi}_2 &= \omega_2. \tag{8}
\end{align*}
\]

Averaging Eq. (7) using Eq. (6) yields two nontrivial one-dimensional coupling functions $q_{1,1} = \frac{\varepsilon}{\pi} \sin(\phi_2 - \phi_1)$ and $q_{2,1} = \frac{\varepsilon}{\pi} \sin(\phi_2 - 2\phi_1)$; all other functions $q_{m,n} = 0$. Thus, the averaged description of Eq. (7) predicts locking only when $\omega_1 \approx \omega_2$ and $2\omega_1 \approx \omega_2$, with triangular Arnold tongues. However, the tongues obtained by numerical simulation of the full model (7) differ from the triangular shape when $\omega_2$ is farther from the resonance frequencies, as shown in Figs. 1(a) and 1(b). Furthermore, numerical analysis of Eq. (7) shows many locking regions, as seen in Fig. 1(c), in contrast to predictions of the reduced model. We explore the applications of the two-dimensional method and compare the one- and two-dimensional methods in the following sections.

### III. EXPERIMENTAL SETUP

Experiments were performed on an electrochemical cell consisting of two Ni working electrodes (99.98% pure), a Pt mesh counter-electrode, and Hg/Hg$_2$SO$_4$/K$_2$SO$_4$ (sat) reference electrode, with a 3M H$_2$SO$_4$ electrolyte, shown in Fig. 2(a). The cell was enclosed in a jacketed glass vessel maintained at a temperature of 11°C. An ACM Instruments multichannel potentiostat was used to individually set the electrode potential $V_i$ of each electrode such that the electrodes undergo transpassive dissolution.

A 650 Ω resistor was attached to each electrode, causing the dissolution current to oscillate [20]. The resulting dissolution currents were measured using a zero resistance ammeter attached to a real-time data acquisition system, Fig. 2(a). The
shape of the electrochemical oscillator wave form depends on applied voltage. Smooth oscillations with a natural frequency of about 0.5 Hz are observed at potentials of approximately 1.105 V. Relaxational oscillations with a frequency of about 0.35 Hz are observed around 1.20 V [21]. As the applied voltage of each electrode in the experimental system can be chosen independently, any combination of smooth or relaxation oscillations is possible.

Here, oscillator 1 refers to the more relaxational oscillator \((\nu_{1}=0.405 \text{ Hz})\) with a natural frequency \(\omega_{1}=0.002 \text{ Hz}\), while oscillator 2 \((\nu_{2}=0.479 \text{ Hz})\) refers to the smooth oscillator with a natural frequency \(\omega_{2}=0.005 \text{ Hz}\). The range is due to the slow drift over time of the natural frequencies of the oscillators as an inherent property of the system.

Negligible intrinsic electrical interactions exist between the uncoupled oscillators. The startup or shutdown of an oscillator does not alter the behavior of the second oscillator. Furthermore, the oscillator dynamics have no interdependence when both oscillators are functioning in the uncoupled state.

Interactions were introduced using real-time coupling of the form

\[
\Delta V_{1}(t) = K[k_{1}x_{2}(t-\tau)], \tag{9}
\]

\[
2\varepsilon x_{1}\Delta V_{2}(t) = K[k_{2}x_{1}(t-\tau)], \tag{10}
\]

where \(\Delta V_{1,2}\) are the changes in the circuit potentials of the elements, \(K\) is the fixed overall coupling gain, \(k_{1}\) and \(k_{2}\) are the coupling gains on oscillator 1 and oscillator 2, respectively, such that \(0 \leq k_{i} \leq 1\), and \(\tau\) is the coupling time delay. The scaled potentials of the elements as a function of time \(x_{i}(t)\) are

\[
x_{i}(t) = V_{i}(t) - I_{i}(t)R_{p}, \tag{11}
\]

where \(V_{i}\) are the applied potentials, \(I_{i}\) are the normalized currents, and \(R_{p} = 650 \Omega\) is the channel resistance. Only linear coupling is considered here, with and without time delay [22,23].
phase advance. Then the phase velocities are fit with a two-dimensional Fourier expansion, Eq. (18) in [7] (when the phase space is not well covered, phase velocities are instead fit with a two-dimensional kerneling function). The coupling functions are then further cleansed using the method in Sec. IV, part B in [7]. Numerics demonstrate that satisfactory results can be obtained already after the first cleansing, since the second cleansing is small compared to the first. Note two limitations of the method (see [7] for details): (i) if the coupling function contains a component dependent only on the phase of the driven system, it will be cleansed; and (ii) generally the coupling function contains a constant term which cannot be separated from the natural frequency; this may be done only if several observations with different yet unknown coupling strengths are available. A MATLAB implementation of the techniques employed for the data analysis may be found online [26].

There is more than one way to obtain the one-dimensional coupling functions, $q^{(i)}$. These can be obtained as in Eq. (6), by averaging from the two-dimensional coupling functions $Q^{(i)}$, or they can be obtained directly from the time series [3]. The methods are conceptually equivalent, and yield nearly identical results. The latter is easier to implement numerically, and we use it here to obtain $q^{(i)}$. First, the periods of each oscillation are calculated. The inverse of the period is the average frequency over the oscillation. Next, we calculate the average phase difference over each oscillation. Here we express phase difference as obtained from the genuine phases; phase differences from the protophases distort the coupling function if the oscillators are dissimilar. Finally, $q^{(i)}$ is obtained by fitting the average frequency as a function of phase difference. The fitting can be performed with a Fourier series or a kerneling function.

V. RESULTS

Experiments were performed using the two-oscillator electrochemical system described in Sec. III. Oscillator 1 has a relaxational wave form ($V_1 = 1.180 \text{ V}$) and oscillator 2 has a smooth wave form ($V_2 = 1.105 \text{ V}$). The oscillators are coupled using the form in Eqs. (9) and (10). Phase models of the two oscillators are then reconstructed from the genuine phase time series according to Eq. (2) or the procedure for the one-dimensional reconstruction discussed in Sec. IV.

In order to highlight the advantages of the two-dimensional coupling function, we compare the coupling functions of the one- and two-dimensional models with symmetrical coupling with and without time delay. Figure 5(a) shows the one-dimensional coupling function of oscillator 2 based on phase difference. The phase model of oscillator 1 is obtained in a similar fashion, but is not shown. This coupling function quantifies the oscillator’s average change in frequency over a period. For example, when $\Delta \phi = \pi/2$ the frequency of oscillator 2 increases relative to its natural frequency. Figure 6(a) shows the two-dimensional coupling function of oscillator 2 based on each phase independently. The one-dimensional phase model, Fig. 5(a), is the average of this two-dimensional model over trajectories between two crossings of $\phi_2 = 0$, i.e., one period. Note that the amplitude of the one-dimensional coupling function is an order of magnitude smaller than the two-dimensional due to the averaging. The two-dimensional function provides a mapping between instantaneous changes in phase velocity and the state of each system. For example, oscillator 2 advances most rapidly near $\phi_1 = \pi/2$ and $\phi_2 = 3\pi/2$.

Further experiments were performed in order to quantify the effects of time delay. From the definitions of the coupling function in Eqs. (4) and (6), time delay is expected to shift the stimulation function in the phase of the perturbing oscillator. Figure 5(b) shows the coupling function of oscillator 2 based on phase difference for symmetric coupling and time delay of $\tau = 1.8 \text{ s}$. The phase model based on individual phases for this case is shown in Fig. 6(b). Notice that both coupling functions are translated as expected. However, the two-dimensional coupling function clearly distinguishes between shifts in the two phases: Fig. 6(b) becomes nearly identical to Fig. 6(a).
if one shifts the phase of the forcing oscillators \( \phi_1 \) by \( \pi \phi_2 = 4.58 \text{ rad} \). Therefore, changes in coupling time delay are measurable from the two-dimensional model to within an additive factor of \( 2\pi \). Time delay may be recovered from the one-dimensional model provided that the response function is known to be time invariant.

Now we investigate the effect of changing coupling magnitude on the phase models. Focus is placed on the two-dimensional model, as it provides a more complete description of the system. Oscillator 1 is a relaxational oscillator (\( V_1 = 1.210 \text{ V} \)) with a natural frequency of 0.42 Hz \( \pm \) 0.02 Hz and oscillator 2 is a smooth oscillator (\( V_2 = 1.105 \text{ V} \)) with a natural frequency of 0.53 Hz \( \pm \) 0.01 Hz. Figure 7 shows the coupling functions of oscillators 1 and 2 as a function of the genuine phases. The means of the coupling functions are less than 1% of the natural frequency, indicating a negligible change in average frequency due to coupling. Three coupling combinations \((k_1:k_2)\) are shown: symmetric, asymmetric (1:2), and highly asymmetric (1:10). As expected from Eq. (4), the amplitude of the surface variations decreases with diminishing stimulation magnitude. Also note that the functional dependence of each coupling function on its own phase is characteristic of the oscillator’s response function [19].

The magnitude of an oscillator’s response is quantified by the \( L_2 \) norm of its coupling function, \( \mathcal{Q} \). Figure 8 shows the dependence of the coupling function norms on the coupling strengths \( k_1 \) and \( k_2 \). In this series of experiments the coupling strength to oscillator 2 was held constant at \( k_2 = 1.0 \), while the strength of the coupling to oscillator 1, \( k_1 \), was increased incrementally from zero to 1.0. In a subsequent series of experiments with the same oscillators, the coupling strength on oscillator 1 was held constant at \( k_1 = 1.0 \) and \( k_2 \) was decremented from 1.0 to zero. Figure 8 shows that the norm of the coupling function increases linearly with increasing coupling strength. This experimentally confirms that the coupling of electrochemical oscillators in the range of parameters studied is predominantly described by first-order terms in the coupling strength, i.e., we are in the regime of linear response.

The relative magnitudes of the coupling functions indicate the coupling directionality between the oscillators. In the electrochemical system described above, the values of \( k_1 \) and \( k_2 \), and therefore the relative coupling magnitude, are known from the experimental setup. The applied directionality is

\[
\delta_A = \frac{k_1 - k_2}{k_1 + k_2}.
\]

Thus, \( \delta_A \) varies between \(-1 \) and 1. The more positive the directionality, the stronger \( k_1 \) is relative to \( k_2 \). A directionality of zero indicates equal coupling strengths, i.e., \( k_1 = k_2 \). We can compare this quantity with the observed directionality, defined as

\[
\delta_O = \frac{C_1 - C_2}{C_1 + C_2},
\]

where \( C_i = ||Q^{(i)}|| / \omega_i \). As shown in Fig. 8, the relaxational oscillator has a greater response to the same coupling gain and thus less voltage perturbation, according to Eq. (9). Therefore \( \delta_A \) and \( \delta_O \) are generally different. When the norms are scaled

FIG. 7. (Top row) \( Q^{(1)}(\phi_1,\phi_2) \); (bottom row) \( Q^{(2)}(\phi_2,\phi_1) \). \( k_2 = 1.0 \) for all plots. [(a), (d)] \( k_1 = 1.0 \), [(b), (e)] \( k_1 = 0.5 \), and [(c), (f)] \( k_1 = 0.1 \).

FIG. 8. (a) Relaxational oscillator: Norm versus gain \( k_1 \), linear fit: \( ||Q^{(1)}|| = 0.045 k_1 + 0.003, R^2 = 0.998 \). (b) Smooth oscillator: Norm versus gain \( k_2 \), linear fit: \( ||Q^{(2)}|| = 0.032 k_2 + 0.001, R^2 = 1.000 \), where \( R^2 \) is the square of the correlation coefficient.
by the response slope from Fig. 8, one restores \( d_O = d_A \).

Here we know \( k_{1,2} \) and thus can obtain the response slopes; in general, \( k_{1,2} \) may not be known.

Next we carried out experiments on an electrochemical oscillator with adjustable harmonic forcing. Figure 9 shows the coupling functions, \( Q^{(1)} \) and \( q^{(1)} \), when the forcing frequency is 5% faster [(a) and (c)], and when the forcing frequency is 5% slower [(b) and (d)] than the natural frequency of the oscillator for a smooth \((V = 1.105 \text{ V})\) oscillator. We see that the two-dimensional coupling function varies with forcing frequency, while the change is not detectable in the one-dimensional coupling function. The observed dependence of the two-dimensional coupling functions on the frequency of the driving has also been observed numerically with the harmonically forced van der Pol and periodic Rössler oscillators.

VI. DISCUSSION

In this paper we evaluate the coupling functions for two coupled nonlinear electrochemical oscillators directly from measured signals. We evaluate both the one- [3,29] and two-dimensional [7] coupling functions and compare the results of the models. While both models may recover changes in coupling time delay, only the two-dimensional model clearly distinguishes between changes in oscillator character and changes in time delay. We show that the relative magnitudes of the coupling functions quantify the directional difference in coupling [27,28]. Using a toy model we show that the two-dimensional coupling function varies with forcing frequency, while the change is not detectable in the one-dimensional coupling function. The observed dependence of the two-dimensional coupling functions on the frequency of the driving has also been observed numerically with the harmonically forced van der Pol and periodic Rössler oscillators.

The effect of forcing frequency on the coupling function

\[ Q^{(1)}(\phi_1, \phi_2) \]

is known to be time invariant, the two-dimensional coupling function is the preferable model for inferring time delay.

We performed experiments in which we varied the ratio of the coupling components, \( k_1 \) and \( k_2 \) in Eqs. (9) and (10). We then calculated the two-dimensional coupling functions. Deviations from linear phase advance indicate points in phase space where an oscillator is susceptible to perturbation and is stimulated. This is nicely visualized in the top row of Fig. 7 which shows the instantaneous frequencies of the relaxation oscillator. There is a dominant ridge in the middle of the surface corresponding to maximum amplitude of the smooth oscillator, and therefore the greatest stimulation. A similar result is seen for the smooth oscillator in Fig. 7(c). The largest instantaneous frequency on the ridge occurs near \( \phi_1 = 3\pi/2 \), which corresponds to the large amplitude in the oscillator phase-dependent response curve [19]. From the coupling functions, the coupling directionality was calculated, as in Eq. (13). The nearly linear increase in the norms of the coupling functions with gain, shown in Fig. 8, is a verification of the two-dimensional reconstruction.

Using the toy model of Eq. (7), we highlight the differences of the predictions of the one- and two-dimensional models. We construct two Arnold tongues, and show that the synchronization gains predicted by the two models differ increasingly as the forcing frequency becomes farther from the resonance condition. Additionally, we show that the two-dimensional model predicts many regions of synchrony in a Devil’s staircase; the one-dimensional model predicts only two regions of synchrony. The differences between the predictions of the two models in a relatively simple and explicitly defined system illustrate how the two models may differ in more complex systems.

Finally, we show from experimental data that the two-dimensional coupling function depends upon the forcing frequency, while the one-dimensional coupling function does not (Fig. 9). As already mentioned, the phase approximation is valid if the cycle is sufficiently stable and therefore the amplitudes can be considered as fixed. For this case the coupling function \( Q^{(1)} \) can be reconstructed from data. If the coupling is sufficiently small, this function can be approximated by only one term of the series equation (3), i.e., \( Q^{(1)}(\phi_1, \phi_2) \approx \epsilon Q^{(1)}(\phi_1, \phi_2) \), and in this approximation the function depends solely on the phases, but not on the frequencies. However, the condition when the first approximation suffices is not yet known, and if it is not fulfilled, we can expect a dependence on the frequency and on the amplitude of the forcing. In this experiment, neither the stimulation function nor the response function changes, so we may infer according to Eq. (4) that the first-order approximation does not hold. The effect of forcing frequency on the coupling function...
was not previously predicted or shown; this effect as well as the range of applicability of the first-order approximation represent opportunities for further study.

An interesting and practically important problem is determination of the response function $Z_i(\phi_i)$ from the observation of the driven system. The ability to separate stimulation and response could be useful in any system where coupling evolves over time, such as systems where learning occurs [30,31]. When the first-order approximation is valid, as in Eq. (4), the coupling function can be represented as a product $Q^{(1)}(\phi_i, \phi_j) = Z_i(\phi_i)h_i(\phi_j)$. Because there is indication of significant higher-order terms in the coupling function, the first-order approximation is not valid here and the deconvolution is not possible. We can suggest an alternative explanation for this; the driving may enter the state-space equations (1) as a multiplicative term, e.g., as $f(x_1)g(\nu t)$; in the process of phase reduction this term yields a function of two phases which cannot be written as a product of two one-dimensional functions. This important issue also requires further studies.

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