First-order synchronization transition in a large population of relaxation oscillators

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Abstract

We study the synchronization transition in a large population of coupled oscillators. In contrast to a continuous non-equilibrium transition for weakly coupled phase oscillators with a unimodal symmetric frequency distribution, we find in experiments with photo-chemically coupled catalytic micro-oscillators a discontinuous synchronization transition with hysteresis. Closer examination reveals that the relaxation character of the oscillators and their strong coupling are responsible for the hard synchronization transition. Accompanying numerical simulations support the hypothesis that the discontinuous character of the transition is structurally stable with respect to changes in network connectivity and modality of the frequency distribution. Due to the prevalence of relaxation oscillators in nature and technology, we expect the discontinuous synchronization transition to play role in diverse systems including neural tissue and genetic regulatory networks.
Main Body

The synchronization of coupled oscillators is a fascinating manifestation of self-organization and is essential to the functioning of a vast range of phenomena in nature and technology (1). Since Christiaan Huygen's discovery of synchronization in pendulum clocks in 1665, self-emergent synchronization of oscillating units has been observed in a plethora of natural systems, including flashing fireflies (2), pedestrians on a bridge locking their gait (3), circadian clocks in the brain (4), or cardiac pacemaker cells (5). In addition, synchronization underpins the dynamics of a variety of artificial systems such as power grids (6), or traffic patterns (7). Today, synchronization is considered a cornerstone in the cooperative behavior of active matter, and many studies in this field have become beacons of inspiration for investigators in complex systems (8, 9).

The process of synchronization can be construed from a statistical mechanics perspective as a non-equilibrium phase transition, where a synchronized state emerges from an incoherent one as the coupling strength is increased. The pioneering works of Winfree (10) and Kuramoto (11) have shown that in populations of weakly coupled phase oscillators, such a transition proceeds continuously and reversible in a second-order fashion. This was also experimentally confirmed by Kiss et al. with electrochemical oscillators (12). Later investigations in this direction have shown that a discontinuous, first-order transition with hysteresis can also occur in certain specific cases within the Kuramoto model (13–15). This latter phenomenon has been hypothesized to play a role in the onset of anesthesia-induced unconsciousness (16), epileptic seizures (17), hypersensitivity in chronic pain (18) or the memory process (19).
**Fig. 1. Coupled photochemical oscillators.** (a) Experimental setup. A thermostatted open reactor hosting \( \sim 2600 \) chemical oscillators is spectrophotometrically monitored in fluorescence light (>550 nm) with a camera. Recorded light intensities \( f_i \) determine the photochemical feedback \( I_i \) which is applied with a spatial light modulator. (b) Camera image of the fluorescing oscillator reservoir. The connectivity between oscillators is overlaid in blue. (c) Hysteresis loop of the Kuramoto order parameter \( R \) in the case of \( N = 1000 \) all-to-all coupled oscillators with normally distributed natural frequencies (standard deviation \( \sigma_\omega = 0.005 \text{ rad/s} \)). See supplementary movie S1.
We studied experimentally the onset of synchronization in large ensembles of coupled relaxation oscillators. By employing the Belousov-Zhabotinsky (BZ) chemical reaction, individual oscillatory units were constructed in the form of ion-exchange resin beads. A photosensitive catalyst ruthenium(II)tris(bipyridine)-chloride is absorbed in these particles which are then immobilized in a small acrylic plate and immersed in a catalyst-free reaction solution. The result is a reservoir of uncoupled chemical micro-oscillators which are neuromorphic in the sense that their intrinsic dynamics behave similar to biological neurons and show an excitable response to externally applied perturbations.

Networks with desired connectivity and natural frequencies can be realized by photo-chemically coupling beads together. This is achieved with the experimental setup presented in Fig. 1a. During an oscillating cycle, the photosensitive catalyst switches periodically between its reduced and oxidized form. The phase of each oscillator is thus monitored by measuring the catalyst concentrations via its fluorescence intensity $f_i$ with a CMOS camera. Using a predefined coupling scheme,

$$I_i = I_0 + K \sum_{j=1}^{N} W_{ij}[f_j(t) - f_i(t)],$$

an individual photochemical feedback $I_i$ is calculated and projected on each micro-oscillator with a spatially modulated light source. The natural frequencies of the beads are measured at the start of each experiment under the uniform background light $I_0$ and then employed in forming populations with arbitrary frequency distributions. Different connectivities can be implemented by choosing an appropriate weighted adjacency matrix $W_{ij}$. Figure 1b depicts the image of the bead array in fluorescence from which the individual intensity values $f_i$ are extracted with the underlying network graph superimposed.
Fig. 2. Experimental observation of hysteresis for bimodal frequency distribution. (a) Time protocol of coupling strength. (b) Time evolution for the instantaneous frequencies of \( N = 200 \) oscillators. The color of each line corresponds to the natural frequency of the nodes, respectively. The oscillators synchronize in-phase at \( K_1 = 0.72 \), but transition back to incoherence at \( K_j = 0.56 < K_1 \). (c-e) Individual fluorescence values for clustering, synchronized and incoherent states during the cycle. Synchronization from incoherent initial conditions (e) proceeds with the formation of phase clusters (c) which delay the onset of synchronization (d) leading to hysteretic behavior. See supplementary movie S2.
For each experiment, the coupling strength $K$ is cycled from low to high values and back. We monitor the onset of synchronization using the Kuramoto order parameter

$$ R = \frac{1}{N} \left| \sum_{j=1}^{N} e^{i\phi_j(t)} \right|_t, $$

where $\phi_j(t)$ represents the phase of the $j$-th oscillator (calculated by linear interpolation between consecutive firing events) and $\langle ... \rangle_t$ denotes time averaging. The order parameter ranges from 0, when the phases are incoherent, to 1, where all the phases align perfectly. The onset of synchronization can be captured from the dependence of the order parameter on the coupling strength. Such an order parameter curve is shown in Fig. 1c in the case of $N = 1000$ globally coupled oscillators with normally distributed natural frequencies (Fig. S1). Upon increasing $K$, there is an abrupt transition to a highly synchronized state at a critical value of the coupling strength, $K^*_1 = 0.4$. Once this phase is formed, it remains stable until $K$ is decreased to a significantly smaller value, $K^*_1 = 0.1$. This is reminiscent for a first-order phase transition, with hysteresis.

The next paradigmatic scenario where the synchronization in networks of weakly all-to-all coupled phase oscillators was extensively studied (19, 20) is the case of a bimodal frequency distribution. The frequency time trace in Fig. 2b displays asymmetry with respect to the coupling strength, indicating hysteretic behavior. The plot shows that the natural frequencies of oscillators are recovered approximately at the end of the experiment, indicating that parameter drift due to aging effects is negligible for the duration of the experiment. At low coupling strengths the fluorescence-time plot in Fig. 2c shows the presence of antiphase clusters ($\alpha$) with intercluster switching (21, 22). Before the onset of global synchronization, the low-frequency subpopulation achieves in-phase synchronization, while the high-
frequency group remains incoherent, but displays an average increase in instantaneous frequency ($\beta$). Once the synchronized state is established at $t \approx 5300 \text{ s} \quad (K_\uparrow = 0.72)$, the population oscillates with the natural frequency of the fastest oscillators ($\gamma$). This regime is characterized by almost perfect phase alignment, with the fast oscillators entraining the entire population (Fig. 2d). The destabilization of the synchronized states at $t \approx 7800 \text{ s} \quad (K_\downarrow = 0.56)$ is mediated by the loss of frequency-coherence of the slower subpopulation ($\delta$). At the end of the experiment, we recover the fully incoherent state that we also observe in the beginning for very low ($K < 0.2$) coupling strengths (Fig. 2e).

To gain more insight in the mechanism of the observed first-order synchronization transition, we performed numerical simulations using an established model of the BZ chemical kinetics (21). Fig. 3 shows the comparison of the hysteretic order parameter cycles between experiments and simulations in the case of globally coupled oscillators with normal and bimodal distributions. In both cases, the ascending branch of the hysteresis cycle is characterized by a persistence of low order parameter values. The detailed inspection of the collective node dynamics in the case of normal (Fig. S2) and bimodal (Fig. 2) frequency distributions, reveals that the abrupt emergence of an in-phase synchronized state is preceded by the formation of antiphase clusters. This effectively suppresses the onset of the former, resulting in hysteretic behavior. The critical coupling strength for in-phase synchronization corresponds to the point where the anti-phase state becomes unstable.
Fig. 3. First order transitions in globally coupled populations. Hysteresis curves for the order parameter with increasing (blue) or decreasing (orange) coupling strength in chemical experiments (a-b) and numerical simulations (c-d). We consider $N = 200$ globally coupled oscillators with normal (a,c) or bimodal (b,d) natural frequency distributions. See supplementary movie S3 and S4.
Moreover, we found that the collective transition can be described by a reduced model of two identical BZ oscillators. Despite its apparent simplicity, quantitative results can be drawn from a bifurcation diagram (Fig. S3). Up to a certain critical coupling strength, both in-phase and anti-phase states coexist. After the critical point the anti-phase state becomes unstable. An estimate for the bifurcation point coupling strength, $K^* = 4.3 \times 10^{-3}$, agrees quantitatively in the case of normally distributed natural frequencies and qualitatively for a bimodal distribution, where the effects of frequency distribution are more pronounced. The peculiarities of the latter case (the sequence of $\alpha$, $\beta$, $\gamma$ and $\delta$ states) are also accurately reproduced by the simulations in the both the order parameter curves and fluorescence intensity plots (Fig S4).

We also investigated the role of network connectivity in determining the nature of the synchronization transition for relaxation oscillators. We considered two paradigmatic random graphs: the scale-free Barabási–Albert (BA) and the Erdős–Rényi (ER) models, where the natural frequency depended linearly on the corresponding node degree. Previous studies within the Kuramoto model (14) showed that explosive synchronization occurs in BA, rather than in ER networks. For relaxation oscillators, there is a discontinuous first-order transition to in-phase synchronization with hysteresis, regardless of the network connectivity (Fig. S5). This strongly suggests that the occurrence of explosive synchronization in the case of relaxation oscillators depends only weakly on the underlying network topology. Moreover, close inspection of the collective node dynamics (Figs. S6 and S7) show the presence of anti-phase clusters suppressing the onset of in-phase synchronization.
**Fig. 4. Relaxation character determines order of the phase transition.** Phase response curves of the FHN model for time scale separation parameter $\epsilon = 3.2$ (a) and $\epsilon = 0.1$ (b). (c) Phase response curves determined from chemical experiments (dots) together with fitted functions for increasing light intensities. Order parameter curves in the case of $N = 200$ globally coupled FHN oscillators with normally distributed natural frequencies for $\epsilon = 3.2$ (d) and $\epsilon = 0.1$ (e). (f) Schematic representation for the mechanism of first-order synchronization via cluster formation: global temporally localized mean-field (Red), phase response curves for generic oscillators from the two clusters (orange and blue traces) for small (continuous) and large (dashed) coupling strengths. The finite extent of the phase response and pulse shapes allows the existence of intra-cluster phase synchronization with inter-cluster phase difference at finite coupling strengths delaying the onset of synchronization.
To validate our hypothesis on the role of the relaxation character we employ the canonical model for relaxation oscillations and neuronal excitability, the FitzHugh-Nagumo (FHN) model (23). Varying the single time scale separation parameter \( \epsilon \) allows for tuning between harmonic and slow-fast relaxation oscillations (Fig. 4). Simultaneously, we observe that the phase response curve evolves from a linear to a nonlinear dependence on the perturbation strength \( A \). While for negligible time scale separation the PRC is roughly sinusoidal (Fig. 4a), for strong time scale separation it is a discontinuous function whose jump point \( \phi^* \) shifts to earlier phases with increasing \( A \) (Fig. 4b). We observe identical behavior for our chemical oscillators (Fig. 4c).

In numerical simulations of a globally coupled network of \( N = 200 \) FHN oscillators we find a continuous transition without hysteresis for vanishing time scale separation and a discontinuous transition with hysteresis for a distinct time scale separation (Figs. 4d,e). In agreement with the experiments, individual dynamics of oscillators in the bistable region reveal antiphase states and in-phase states during the up- and down-sweep, respectively. The mechanism for the hysteretic transition of relaxation oscillators is deeply rooted in the nonlinear behavior of the PRC (Fig. 4f). The amplitude mean field grows with the number of in-phase synchronized oscillators, but its spikes make it sharply localized in time. In addition the constituent oscillators are also only susceptible to perturbations during a certain time interval, whose size depends on the perturbation strength. Taken together, in the highly synchronized state with \( R \approx 1 \), the mean field perturbations are so large, that the susceptible interval of the PRC covers a major portion of the oscillator period. In turn each spike from the mean field successfully triggers the start of a new oscillation cycle in all oscillators. At low \( R \), when two populations oscillating in antiphase emerge, the mean field is weaker and, in turn, the susceptible interval is narrower. Due to the two populations the mean field has a
frequency that is twice that of any oscillator. However, oscillators from one population are effectively blind to the other, because the collective spike of the latter does not fall into the susceptible interval of the first.

Conclusion

Due to the broken linear dependence on perturbation strengths, ensembles of strongly coupled relaxation oscillators cannot be analytically treated with simplified phase models. Despite this difficulty we investigated the collective onset of synchronization in relaxation oscillators with time scale separation experimentally and numerically. Remarkably, the type of transition is much more sensitive to the relaxation character - as quantified by the time scale separation - than frequency distribution or network connectivity, which is in stark contrast to phase oscillators. Based on our analysis of the underlying mechanism we expect that it plays a role in further ensembles oscillatory systems, such as laser arrays for optical communication and neuromorphic computation (24), and neural networks that can be probed by optogenetic means (25).

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