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Experimental evidence of explosive synchronization in mercury beating-heart oscillators

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We report experimental evidence of explosive synchronization in coupled chemo-mechanical systems, namely in mercury beating-heart (MBH) oscillators. Connecting four MBH oscillators in a star network configuration and setting natural frequencies of each oscillator in proportion to the number of its links, a gradual increase of the coupling strength results in an abrupt and irreversible (first-order-like) transition from the system's unordered to ordered phase. On its turn, such a transition indicates the emergence of a bistable regime wherein coexisting states can be experimentally revealed. Finally, we prove how such a regime allows an experimental implementation of magneticlike states of synchronization, by the use of an external signal.

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I. INTRODUCTION

Synchronization of interacting nonlinear oscillators is possibly one of the most relevant and common phenomena encountered in natural and man-made systems [1,2]. The seminal works describing it [3,4] have, indeed, found a huge body of applications and experimental proofs in a variety of fields ranging, for instance, from biology and ecology to semiconductor lasers and nonlinear optics, to chemical oscillators and electronic systems [2,5,6].

An issue of the fundamental importance is characterizing the nature and properties of the transition between the unordered and ordered (synchronized) phase of populations of networked oscillators, as a control parameter is gradually increased. Recently, it has been shown that the transition to the graph's synchronous evolution may display either a reversible, or an irreversible character. The former case corresponds to a second-order phase transition, and the latter, instead, to a discontinuous transition called explosive synchronization (ES). ES has rapidly become a subject of enormous interest: its presence in networked oscillators has been largely theoretically studied, its root on specific arrangements in the microscale of the network's topology thoroughly discussed, and its occurrence numerically described in several cases and circumstances, including periodic and chaotic oscillators [7–16]. Yet, however, experimental evidence of ES was limited only to analog simulations with electronic circuits [9].

In this article, we give evidence of ES in a laboratory system: a networked ensemble of mercury beating-heart (MBH) oscillators. In particular, we prepare a star network configuration, wherein the natural frequency of each oscillator is chosen in proportion to its network's degree, and show that, as the coupling strength is varied from lower to higher (forward direction) and higher to lower (backward direction) values, the forward and backward curves of the synchronization order parameter do not overlap, thus indicating the existence of a hysteretic region, where the two phases actually coexist. Further, we explain how such an emergent bistability allows the experimental construction of magneticlike states of synchronization. For this latter purpose, we first adjust the coupling strength until the experimental system enters the hysteresis domain with the oscillators staying in the unordered phase, and then an external signal is momentarily applied that entrains all the oscillators, thus shifting the system to its ordered phase. Upon removal of the external signal, the oscillators remain in the synchronized phase (in analogy with the case of a permanent magnet), this way validating experimentally the coexistence of the two system's phases.

II. EXPERIMENTAL DETAILS

The schematic diagram of the used setup is illustrated in Fig. 1. Four MBH oscillators (S_1, S_2, S_3, and S_4) are connected in a network star configuration, with system S_1 being the central oscillator. A solution of 2 M H_2SO_4 is used for all the MBH oscillators, while a concentration of $\geqslant 0.12$ mM for the oxidizing agent $K_2Cr_2O_7$ is used for the *leaves* oscillators S_2, S_3, and S_4. For all details about the arising nonlinear dynamics, including the basic underlying chemistry, the reader is here addressed to the relevant literature which can be found in Refs. [17–19].

For the purpose of our study, oscillator S_1 needs to be kept at a frequency consistently higher than that of the leaves oscillators, in a way that a positive correlation is set between the frequency of each MBH oscillator and its network's degree [7]. This is actually obtained by enforcing various concurrent strategies, i.e., (i) by maintaining the concentration of $K_2Cr_2O_7$ ($\geqslant 0.16$ mM) in S_1 at a higher level than that used for the surrounding oscillators; (ii) by imposing a radius of curvature of the watch glass for S_1 ($=5.56$ cm) in a way that it is smaller than that of the surrounding oscillators ($=8.79$ cm); (iii) by using a volume of mercury in S_1 of 0.7 ml, whereas all the other MBH oscillators are prepared with 3.0 ml volume of Hg. The result of these experimental settings is that the natural frequency of oscillator S_1 (of the surrounding leaves oscillators) is maintained at 10.90 ± 0.05 Hz (5.80 ± 0.08 Hz).

Platinum wires are used to make inert the contact with the mercury drops in each of the systems. The Pt wires are covered with Parafilm in a way that only the small tip is left exposed. The small tip is subsequently submerged in the center of the mercury drop, and therefore it is ensured that there is no contact

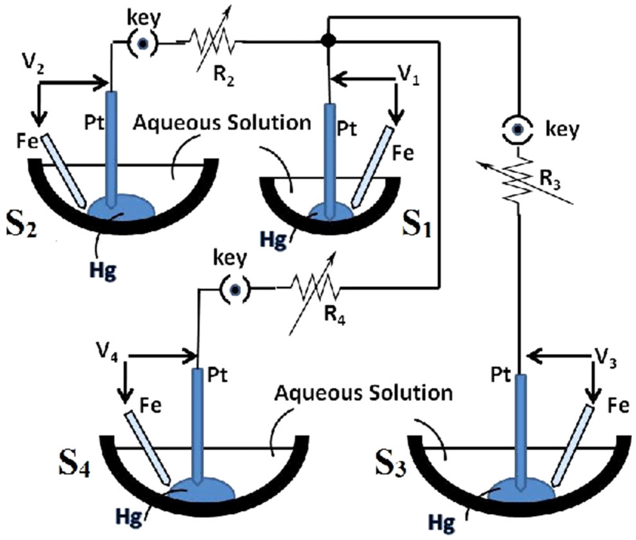

FIG. 1. (Color online) The experimental setup. Schematic diagram of the four MBH oscillators, S_1, S_2, S_3, and S_4, which are connected in a star configuration with oscillator S_1 at the center. The oscillators S_2, S_3, and S_4 are connected to S_1 via variable resistors R_2, R_3 and R_4, respectively. The value of all the resistors is kept identical, i.e., $R_2 = R_3 = R_4 = R$. The iron nails represented by Fe are connected to each other externally using the insulated copper wire. The potential of each of the MBH oscillators (V_i, $i = 1,2,3,4$) is measured with respect to the Fe nails which act as the common ground.

of the tip with the solution. An iron nail of 0.5 mm diameter of 99.9% purity is used for the experiments. The time series of the redox voltages (V_1, V_2, V_3, V_4) of the MBH oscillators are recorded using a digital oscilloscope (Tektronix, DPO 3014). All voltages are measured with respect to Fe nails, as shown in Fig. 1. Furthermore, all Fe nails are electrically connected with the insulated copper wire externally. Box potentiometers (R_2, R_3, R_4 with 2% error) are used for varying the coupling strengths (actually, in our experiment each resistor is kept at the same value, from here on denoted by R). The coupling strength is inversely proportional to R, i.e., if $R = X\ \Omega$, then the coupling strength is $k = 1/X$ mho. A function generator (Tektronix, AF 3021B) is further used to generate the external signal that will be applied to the signal in the second phase of the experiments.

Prior to collecting data, trial experiments were performed wherein the oscillatory behavior of the MBH systems was monitored in a coupling domain from $R = 1$ to $R = 100\ \Omega$ (i.e., the domain which we subsequently used in the experiments). The trial experiments enabled us to fix the position of the Fe nail precisely, i.e., ensuring that there is no permanent loss of contact between the mercury drops and the iron nail, that would lead to the suppression of the oscillatory behavior and that, therefore, needs to be precluded. Eventually, the resistances were varied systematically from higher to lower (forward direction in the coupling strength) and lower to higher (backward direction in the coupling strength) in steps of 5 Ω each, and the resulting time series were recorded at every resistance value in both directions.

III. RESULTS AND DISCUSSION

After time series V_1, V_2, V_3, and V_4 are acquired, the associated instantaneous phases for each one of the four MBH oscillators are evaluated as follows. First, we calculate the Hilbert transform [4,20]:

$$H_i(V_i)(t) = \frac{1}{\pi}\text{P.V.}\int_{-\infty}^{\infty}\frac{V_i(\tau)}{t-\tau}\,d\tau, \tag{1}$$

where P.V. stands for the principal value, and $i = 1,2,3,4$. The instantaneous phases $Z_i(t)$ are then obtained as

$$Z_i(t) = \tan^{-1}\frac{H_i(V_i)(t)}{V_i(t)}. \tag{2}$$

Consequently, the extent of synchronization (the order parameter) in our system is $r = \langle r(t)\rangle_t$, where $\langle r(t)\rangle_t$ is the average over a sufficiently long time window (for our calculation the average is made over a time lapse of 20 s), and

$$r(t) = \frac{1}{N}\left|\sum_{i=1}^{N} e^{\iota Z_i(t)}\right|. \tag{3}$$

The order parameter r lies between 0 and 1. Theoretically, $r \sim 1$ in the fully synchronized state, whereas in the limit $N \to \infty$ one has $r \sim 0$ (actually, $r \sim \frac{1}{\sqrt{N}}$) for the nonsynchronized state. However, since in our system $N = 4$, authors are well aware that r can never be small, and that one has to expect values of the order parameter always above a baseline of the order of 1/2.

For the sake of illustration, we report in Fig. 2 (Fig. 3) the time series of the redox voltages V_1, V_2, V_3, and V_4 when the star configuration of the MBH oscillators is prepared with $R = 100\ \Omega$ ($R = 10\ \Omega$). It is easily seen that the weak coupling configuration at $R = 100\ \Omega$ (Fig. 2) corresponds to the system in its incoherent phase, with all oscillators having independent instantaneous phases. At variance, Fig. 3 reports the voltage time series in the strong coupling case ($R = 10\ \Omega$), where

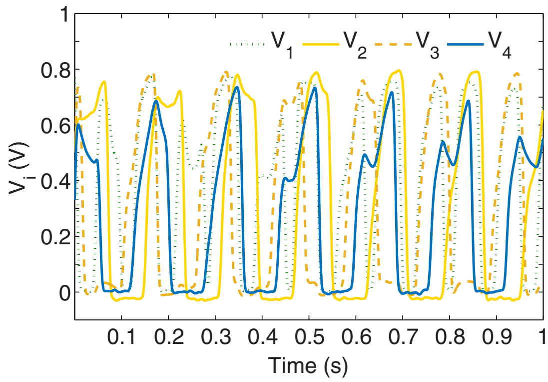

FIG. 2. (Color online) The system's unordered phase. Superimposed time series of redox potentials for the four MBH oscillators [V_1 dotted green (dotted gray), V_2 yellow (light gray), V_3 dashed yellow (dashed gray), and V_4 blue (dark gray) lines] at $R = 100\ \Omega$ (i.e., in the regime of weak coupling strengths). This corresponds to the typical evolution of the systems in the regime of low values for the order parameter r. The time series of V_1 oscillates at its natural, higher, frequency.

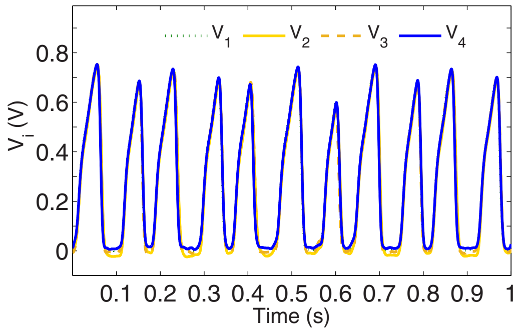

FIG. 3. (Color online) The system's ordered phase. Superimposed time series of redox potentials for the four MBH oscillators [V_1 dotted green (dotted gray), V_2 yellow (light gray), V_3 dashed yellow (dashed gray), and V_4 blue (dark gray) lines] at $R = 10\ \Omega$ (i.e., in the regime of high coupling strength). The systems' dynamics evolve in the ordered (synchronized) phase, and all oscillators lock to the high frequency of the central one.

the system sets in its coherent (synchronized) dynamics. It is observed experimentally that in such a latter situation all the units lock to the high frequency of the central oscillator.

The full quantitative description of the transition between these two phases is shown in Fig. 4, where r versus $1/R$ is reported along the path to synchronization, i.e., when R is gradually and monotonically decreased [forward direction, blue (gray) lines in the figure] and increased [backward direction, dotted blue (dotted gray) curves in the figure] in small steps $\Delta R = 5\ \Omega$. Our experimental evidence points out that the two transition curves have different critical points, and they *do not follow* the same paths. This fact marks the irreversible nature of the transition to synchronization, which is here characterized by a hysteresis behavior pointing to a region of coexistence of the two system's phases.

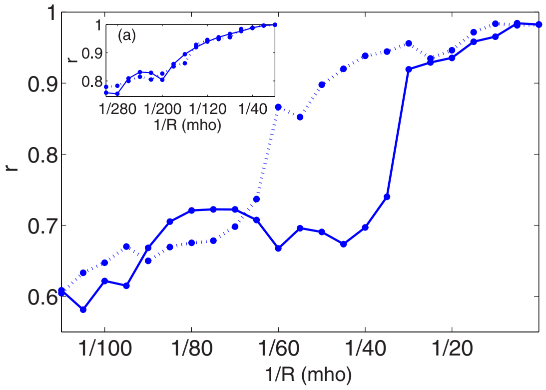

FIG. 4. (Color online) The explosive transition. Order parameter (r) versus the coupling strength ($1/R$). In the experiment, R was varied systematically in steps of $5\ \Omega$. The forward direction, shown in blue (gray) color, and the backward direction, shown in dotted blue (dotted gray) color, correspond to a gradual increase and decrease in the coupling strength. The experimentally observed hysteretic region extends from $1/65$ mho to $1/35$ mho. Inset: The natural frequency of the central oscillator (of the leaves oscillators) is 10.90 ± 0.05 Hz (5.80 ± 0.08 Hz). For the sake of comparison, the inset reports the same forward and backward transitions in a completely different experimental arrangement, where all oscillators are prepared to have quasi-identical natural frequencies. In the inset, R varies between 300 and $5\ \Omega$, with a step size of $20\ \Omega$; the central oscillator has a frequency of 7.725 Hz; while the frequencies of the surrounding oscillators are, respectively, 7.563, 7.838, and 7.65 Hz. The volume of the Hg is kept at 2 ml, and the concentration of $K_2Cr_2O_7$ (in oscillator 2 M H_2SO_4) was maintained at 0.1 mM. It is evident that the transition root to the synchronized phase has, here, a smooth and fully reversible nature.

In order to highlight the crucial role played by the microscopic imposed correlation between oscillators' natural frequencies and network's degrees, we have also carried out trials in a completely different experimental arrangement, wherein all oscillators, including the central one, are engineered to display almost the same frequency. The results are reported in the inset of Fig. 4, and the new experimental specifications are reported in the caption of the same figure. It is clear that these other experimental conditions determine a completely different situation, where the transition to synchronization is instead reversible and second-order-like, i.e., both the forward and backward transition curves perfectly overlap, with a loss of the hysteresis domain.

It is crucial to remark that the experiments conducted here are extremely hard to perform in a fully reproducible way. The MBH system is indeed a chemomechanical oscillator exhibiting both mechanical (varying radii of the mercury drops) and chemical oscillations (redox). The concentration of the oxidizing agent in the solution, the accuracy of the positioning of the Fe nail with respect to the mercury drop, and the curvature of the vessel (watch glass) are all crucial variables, and any variation or fluctuations of those parameters might determine significant changes in the experimental observations. For instance, in certain experiments, a cessation of the oscillatory behavior can even be observed due to a permanent loss of contact between the Fe nail and the Hg drop. Furthermore, as the concentration of the oxidizing agent unavoidably diminishes in time, a drifting of the experimental conditions is actually fated, hampering the exact reproducibility of the experiment. Nonetheless, while all these inherent complications may result in hysteresis plots from different experiments displaying nonidentical critical points (with possibly both the critical coupling strengths for the forward and backward transition curves and the amplitude of the jump in the order parameter being slightly different from one experimental run to another), the presence of such critical points and the emergence of the hysteresis domain is always observed. Therefore, although for the absolute values of the transition points, the profile presented in Fig. 4(a) persists consistently, giving confidence that the experimental results presented here are actually robust, and the underlying mechanisms generic.

Finally, we implement the strategy introduced in Ref. [10], which exploits the emergent bistability for the experimental production of magneticlike states of synchronization. To that purpose, the experiment is first prepared within the hysteresis domain (actually, we run two trials at two different values of R: 70 and 40 Ω) with the systems' dynamics being in the unordered phase. Then, an external signal (consisting in a square wave signal with a peak to peak amplitude of 2 V) is momentarily added using the relay keys sketched in Fig. 1, which has the effect of entraining frequencies and

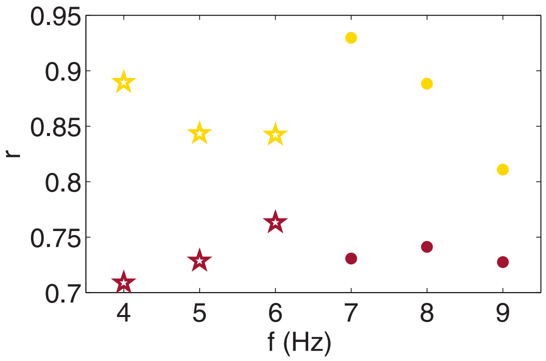

FIG. 5. (Color online) Magneticlike states of synchronization. Scattered plot of the order parameter r versus the external signal frequency f (simultaneously superimposed to all oscillators; see text for details on the procedure). The external signals are applied inside the hysteretic region of Fig. 4, and removed after a while. Dots and stars stand, in fact, for two different values of coupling strength (stars for 1/70 mho and dots for 1/40 mho). Red (dark gray) symbols represent the initial state of the r and yellow (light gray) symbols represent the final state of the r, before the application of the external signal and after the external signal is removed.

instantaneous phases of all the oscillators, thus shifting the system to its ordered phase. Upon removal of the external signal, we consistently observe that the oscillators actually remain in their state of synchronization.

The results are reported in Fig. 5. The red (dark gray) symbols stand for the initial values of the order parameter (before the supply of external signal), while the yellow (light gray) symbols mark the r values measured after the removal of the external signal. The robustness of such an observation is checked by changing the frequency f of the external signal. In particular, signals with frequencies 4, 5, and 6 Hz were superimposed to the MBH oscillators at a coupling strength of 1/70 mho (stars in Fig. 5), while the system's state corresponding to a coupling strength 1/40 mho was successfully manipulated with frequencies of the external signal of 7, 8, and 9 Hz (dots in Fig. 5).

IV. SUMMARY

In conclusion, here we provided experimental evidence of explosive synchronization in a laboratory system. Using four mercury beating-heart oscillators coupled via resistors and placed in a star configuration, we have shown that the system possesses two distinct phases, the unordered and the ordered (synchronized) one, and that the transition between these phases is irreversible, abrupt, and associated with the presence of a hysteretic region where such phases coexist. Furthermore, we gave evidence that the dynamics of our chemomechanical oscillators can be conveniently and permanently switched from one phase to the other when the experiment is prepared within the hysteretic (bistable) region, and when an external signal is momentarily superimposed. The robustness of our experimental findings suggests that, under appropriate conditions, explosive synchronization should be observed, actually, in other numerous and diverse physical systems.

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