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Control, synchronization, and replicability of aperiodic spike trains

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Aperiodic spike sequences, characterized by an almost constant spike amplitude and randomly distributed interspike intervals, are studied experimentally in an electrochemical cell. In the first set of experiments, these aperiodic spike trains are converted to regular spike sequences using periodic forcing. Subsequently, synchronization of two such irregular spike time series is achieved for an appropriate bidirectional coupling. Finally, reproducibility of these irregular spike profiles is evoked by virtue of an externally superimposed stochastic stimulus.

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Spiking behavior has been observed in a variety of chemical [1], optical [2,3], and biological systems [4–6]. In chemical processes, an interplay of excitatory and inhibitory states of the system results in the induction of spike sequences with regular and/or irregular interspike intervals. Mathematically, this inception of spiking behavior is often attributed to the topology of the nullclines for the governing dynamical equations. The shape of these nullclines and their mutual intersections determine the features of the emergent relaxation oscillations. Since numerous physiological processes are controlled by underlying chemical reactions, spiking is observed in various biological timeseries such as ECG (electrocardiogram), EEG (electroencephalogram), etc. In particular, aperiodic spiking behavior is the signature dynamics of the neuronal response measured from the axons [7,8]. The ubiquitousness and relevance of these spike sequences to physiological rhythms makes them a topic of current interest. Analyzing and manipulating spiking behavior in the framework of nonlinear dynamics is important due to its possible pertinence to biological systems. For example, complete and partial synchronicity of irregular spiking in coupled neuronal oscillators apart from having far-reaching implications in neuronal and cognitive sciences [9–11] could have possible medical relevance to neurobiological systems. Moreover, enhanced reliability of firing sequences under the influence of an external noise [12,13] is deemed important in elucidating the mechanism for coincidence detection.

In the present work, we experimentally studied the irregular spike sequences produced by an electrochemical oscillator. Initially, a parameter space exploration was carried out to locate the irregular (chaotic) dynamics of interest. Subsequent to characterizing the nature of the chaotic spiking, a series of different experiments were carried out subjecting the autonomous spike sequence to control, synchronization, and superimposed external noise. In the first series of experiments, using a forcing technique, these chaotic spikes were converted to periodic oscillations, thereby rendering the successive interspike intervals constant. Second, we designed an electrochemical cell in which two such chaotic oscillators (each exhibiting irregular spiking) were subjected to a mutual coupling. This, for appropriate values of the coupling parameters, resulted in the inception of amplitude synchronization between the two irregular spike sequences. Finally, the irregular spiking behavior was subjected to superimposed external noise. This stochastic forcing induced replicability in the observed spike sequences. Consequently, for successive experimental runs in the presence of the superimposed noise, identical irregular spike sequences were obtained.

A standard three-electrode electrochemical cell was designed to study the aperiodic spike sequences experimentally. The cell was operated potentiostatically such that the anodic potential (V) between the anode and reference electrodes was maintained constant and at the same time the anodic current (I) between the anode and the cathode was monitored. Therefore, in these experiments, V was the control parameter while I was the system observable. The anode was made of pure iron (Aldrich, 99.98% purity) disk with a diameter of 6.3 mm shrouded by epoxy. The cathode was a 6.3 mm graphite bar, and the reference was a saturated calomel electrode (SCE). The electrolyte solution was a mixture of 1.0 M sulfuric acid, 0.4 M potassium sulfate, and 13.41 Mm of potassium chloride. A volume of 300 ml was maintained in the cell. The electrochemical cell was operated at room temperature (≈ 25 °C). The dominant reaction occurring in this electrochemical setup was the anodic dissolution of iron in the acidic media [14,15]. Since the anode was shrouded by epoxy, it was ensured that the anodic reactions (dissolution) were restricted to the surface of the anode exposed to the electrolytic solution. It needs to be mentioned that the depth of the iron disk (anode) in the electrolytic mixture affected the spiking profile. The appropriate autonomous dynamics (aperiodic spiking) was located using the standard cyclic voltammogram method [16]. This method involved incrementing and decrementing the control parameter (V) at a predetermined velocity and recording the corresponding behavior of the system observable (I). The cyclic voltammogram technique, albeit not as accurate as a bifurcation diagram, was extremely effective in mapping out the different dynamical behaviors in the parameter domain.

Figure 1(a) shows a section of the I timeseries for the autonomous dynamics. The system parameters are provided in the corresponding figure caption. The oscillatory behavior observed is characterized by spikes of almost constant amplitude and irregular interspike intervals. Figure 1(b) shows the autocorrelation function (ACF) calculated for the time series (longer section) in Fig. 1(a). The fact that ACF goes to zero is indicative of chaotic spiking dynamics. A histogram calculated for the elapsed time between successive peaks (interspike intervals) is shown in Fig. 1(c). The Fourier spectra for the time series is computed and provided in Fig. 1(d). This multiple peak histogram in conjunction with the broad-

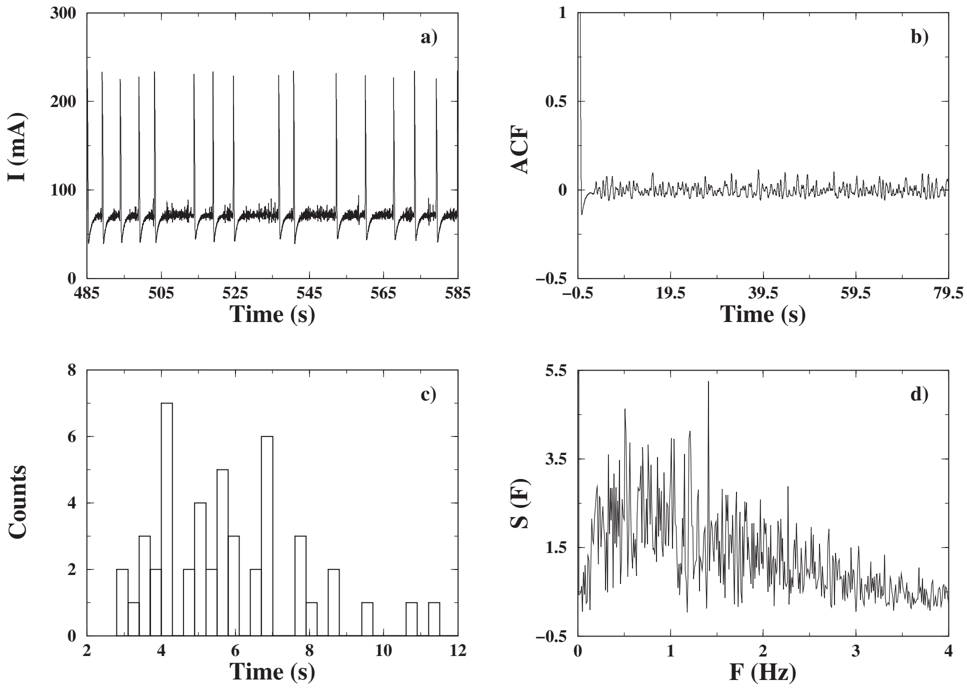

FIG. 1. (a) The autonomous time series of anodic current (I) showing aperiodic spiking behavior. The anodic voltage (V) was fixed at 640 mV. In all the experiments involving three electrodes the anode, cathode, and reference were placed in a triangular configuration. The electrolyte composition is the same as mentioned in the text. (b) The autocorrelation function (ACF) calculated for a larger segment of the time series presented in (a). (c) The multipeaked histogram for the interspike intervals obtained from the time series (larger segment) of (a). (d) The broadband Fourier spectra computed for the time series (larger segment) of (a).

band Fourier spectra indicates that the autonomous dynamics exhibits chaotic spiking behavior.

In the first phase of experiments involving aperiodic spike sequences, we tried to convert the autonomous chaotic spiking to a periodic spike train using a non-feedback-control technique [17,18]. This technique is based on periodic modulations of the control parameter V. Under this control strategy the anodic voltage (V) is continuously perturbed such that

$$V(t) = V_0 + \gamma \sin(2\pi\nu t), \tag{1}$$

where V_0 corresponds to the set point of the autonomous system and $\gamma \sin(2\pi\nu t)$ yields the sinusoidal modulation of the control parameter. γ and ν are the forcing amplitude and the forcing frequency of the control term.

The forcing amplitude (γ) was chosen such that the maximum perturbations were about 10% of the base value of the set point (V_0). The forcing frequency ν was calculated in the following manner: The successive interspike intervals of the autonomous dynamics were analyzed. Subsequently, an average of this irregular distribution of interspike intervals was computed. This average time period of the chaotic spikes, when converted to frequency, yielded the first estimate of the forcing frequency. Thereafter, this forcing frequency was fine tuned online (while the perturbations were being implemented) until optimal control was achieved.

Figure 2(a) displays the anodic current (I) time series with alternating regions of dynamics with "control off" and "control on." Figure 2(b) shows a blowup from a section without

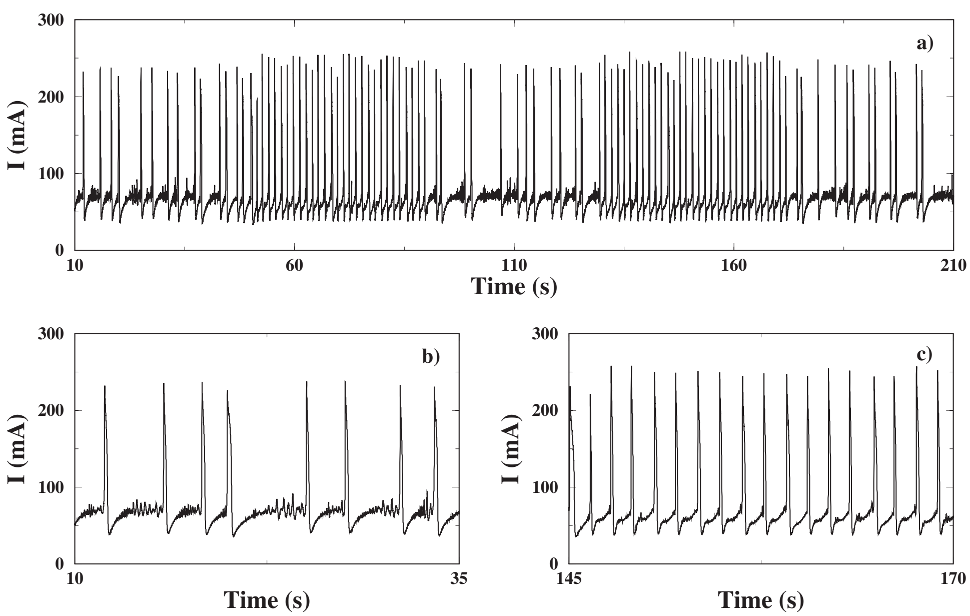

FIG. 2. (a) Control of period-1 spiking behavior in the electrochemical cell exhibiting chaotic spiking behavior. The anodic potential (V_0) was 640 mV. The electrolyte composition is the same as mentioned in the text. The anodic current is plotted over a period during which the control is switched off, on, off, on, and off. The control parameters are γ =64 mV and ν=0.4 Hz. (b) A close-up of the domain without control shows the autonomous aperiodic spike sequence. (c) A close-up of the domain with control shows the controlled period-1 spiking behavior.

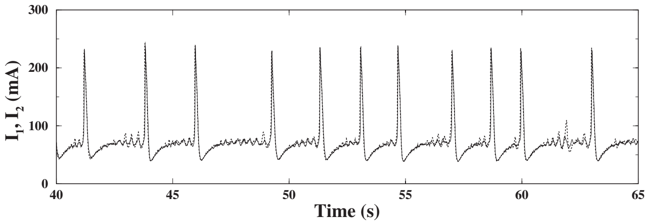

FIG. 3. The superimposed chaotic anodic current (I_1, I_2) time series reveal the emergence of the amplitude synchronization phenomena when the two anodes were placed at a horizontal distance of 1.5 cm. The two anodes and the cathode were placed in an isosceles triangle configuration wherein the vertical distance between the cathode and the anodes was 2.5 cm. The two anodic voltages (V_1, V_2) were chosen identical at 640 mV. The electrolyte composition is the same as mentioned in the text.

control. It shows the aperiodic nature of the spiking in the absence of the forcing term. Figure 2(c) shows a blowup from a section with control. It reveals that under the influence of appropriate forcing, the anodic current (I) time series exhibits a regular (period-1) spiking profile. The effectiveness of the control was sensitive to the choice of the control parameters (γ and ν). In particular, there was a band of resonant frequencies for which control of aperiodic spiking was easier.

In the second set of experiments, chaotic synchronization of these aperiodic spike sequences was considered. Chaotic synchronization, although discovered earlier [19–21], was brought to the forefront by the pioneering work of Pecora and Carroll [22]. This phenomenon has subsequently been observed in numerous physical, chemical, optical, and biological processes [23–29]. Synchronization of spike trains is deemed important due to its direct overlap with the synchronization phenomena observed in biological rhythms [30]. To study the chaotic synchronization of two oscillators exhibiting chaotic spiking, we modified the electrochemical cell used in the control experiments. An extra anode (iron disk) was added and, therefore, the location of the reference and the cathode was adjusted according to symmetry requirements. The rest of the cell characteristics (electrolyte concentration) remained unchanged. The two anodes (iron disks) were immersed in the electrolyte solution facing each other. The only medium of coupling and communication between the two oscillators (anodes) was the electrolyte solution via the possible movements of ions due to migration and/or diffusion [15]. Therefore, the horizontal distance between the two anodes determined the strength of their mutual (bidirectional) coupling. More specifically, the coupling strength was inversely proportional to the horizontal distance between the two oscillators.

Figure 3 shows the two time series (solid and dashed lines) of chaotic spiking superimposed. The cell and coupling parameters are provided in the associated figure caption. These time series indicate that the two chaotic spike trains exhibit amplitude synchronization. Moreover, if one modified the spatial arrangement of the electrodes, other manifestations (lag, intermittent lag, and intermittent amplitude) of the synchronization phenomena were also observed (results not shown). To reiterate, robust amplitude synchronization of the aperiodic spike sequences was observed for appropriate values of the coupling parameters.

Finally, the reliability of these aperiodic sequences under the influence of superimposed stochastic fluctuations was explored. For these experiments, the cell configuration was reverted back to the setup used for the earlier experiments involving control. Previous works studying the interaction of oscillatory dynamics with stochastic perturbations reported that common noise can play a positive role in augmenting the predictability of these dynamics [12,13]. We wanted to verify these results for the aperiodic spike sequences in our electrochemical cell. In the first part, the cell parameters were chosen such that the autonomous dynamics exhibited chaotic spiking behavior. This chaotic spiking was recorded after discarding the initial transient dynamics. Thereafter, the experiment was stopped, the electrode surface polished, and the electrochemical cell started once again to record a second time series (after discarding the transients) of chaotic spiking. This procedure was repeated until nine time series of aperiodic spiking were recorded. In the second part of experiments, to study the interaction of external noise with aperiodic spiking behavior, the control parameter V was modulated such that

$$V = V_0 + \xi, \tag{2}$$

where ξ is the stochastic fluctuations of a predetermined amplitude and whose properties are consistent with that of Gaussian white noise. Analogous to experiments with autonomous dynamics, nine time series were collected (subsequent to discarding the transients) by shutting off the experiments, polishing the anode, and starting again the electrochemical cell. However, in contrast to experiments with autonomous dynamics, in these experimental runs the anodic voltage (V) was being perturbed continuously by an identical noise (ξ) sequence.

Figure 4(a) shows the nine time series superimposed for the repeated (successive) experiments without perturbations. On an average about 22 spikes were recorded for each experimental run. Since initiating and stopping experiments is analogous to starting the dynamical system at different initial conditions, the underlying chaotic nature of the spike trains provokes a total loss of correlation between the spike sequences from distinct experimental runs. Consequently, the time series of Fig. 4(a) exhibits a random distribution of spikes without any mutual correspondence. Under the influence of stochastic modulations, the spiking frequency is obviously enhanced [13]. However, as shown in Fig. 4(b) a strong coincidence between the spikes from different experimental runs (nine) emerges. In other words, mutual correspondence between the spike profiles from successive experimental runs is provoked by the superimposed stochastic stimuli. There exists a minimum (threshold) amplitude of noise beyond which this reported mutual correspondence is observed. These stochastic fluctuations ξ are presented in the lower part of Fig. 4(b). It is remarkable that when subjected to stochastic perturbations the observed spike sequence, albeit irregular, becomes reproducible from one experimental run to the other.

In conclusion, our results show that an aperiodic spike sequence can be manipulated using the techniques of nonlin-

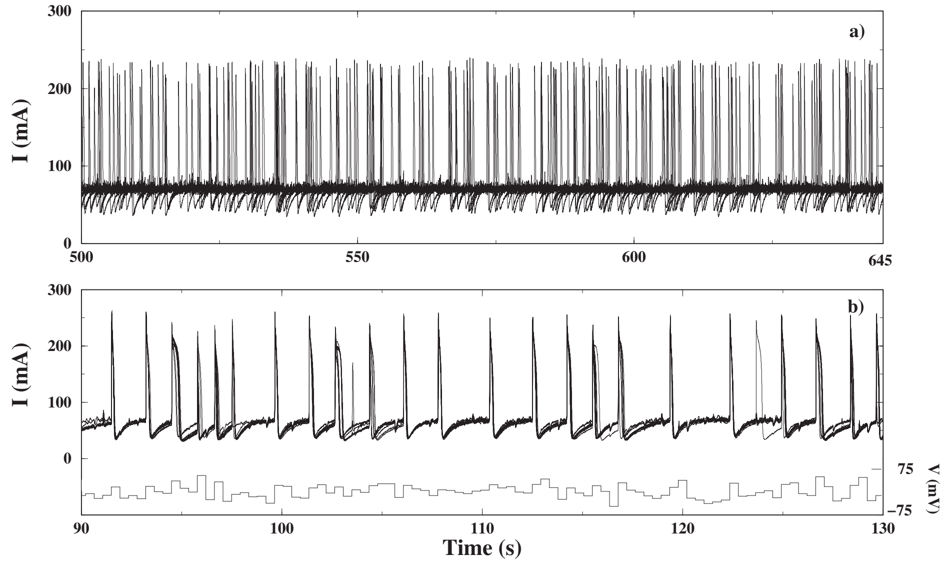

FIG. 4. (a) The superimposed time series (nine) of autonomous chaotic spiking behavior obtained from successive experimental runs. An average of about 22 peaks was recorded for each experiment. The lack of mutual correspondence between these time series is manifested by a random distribution of the observed spikes. The anodic voltage was chosen to be 640 mV. The electrolyte composition is the same as mentioned in the text. (b) The superimposed time series (nine) for the stochastically perturbed chaotic spiking behavior. An average of about 22 peaks was recorded for each experiment. Under the influence of noise the average spiking frequency is augmented. Nonetheless, the spike sequence retains its chaotic profile. The reliability of the spiking sequence under stochastic modulations is manifested by the coincidence of spiking events for the different time series. The anodic voltage was being continuously modulated $V=V_0+\xi$ where $V_0=640$ mV and ξ is the noise sequence provided in the lower part of the figure. The electrolyte composition is the same as mentioned in the text.

ear dynamics. Using appropriate strategies, control, synchronization, and reproducibility of these chaotic spikes trains is achieved. The small imperfections in the obtained experimental results can be attributed to the significant levels of internal noise prevalent in our electrochemical cell. This internal noise in conjunction with the system's drift made the experiments challenging. Since the autonomous dynamics of our electrochemical cell shows striking similarities to the time series recorded from numerous physiological rhythms, the possibility of success for nonlinear techniques in biological systems seems plausible.

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