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Feedback-controlled forcing of meandering spiral waves in an open gel reactor

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The control of spiral wave dynamics has attracted much interest recently. Besides external forcing at a fixed frequency, feedback-controlled forcing has been studied. One of the simplest feedback schemes is to measure the activity level at a particular point of the medium continuously and to apply a spatially uniform pulsatory modulation if a certain threshold is reached at the detector. We have realized this feedback loop using an open gel reactor for the light-sensitive Belousov-Zhabotinskii medium. This allows us to maintain stationary non-equilibrium conditions over several hundred rotation periods of the unperturbed spiral wave. By varying the distance between the detector and the spiral tip, we find different stable branches of the resonance attractor for the same medium. We confirm the constant spacing between neighboring stable branches as predicted by Zykov and Karma and measure the dependence of the resonance drift on the forcing amplitude.

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Spiral waves are among the most prominent examples of spontaneous formation of spatiotemporal patterns in macroscopic systems driven far from thermodynamic equilibrium. They occur as rotating waves of chemical activity in the Belousov-Zhabotinskii (BZ) reaction [1–3], as the coverage pattern on Pt single crystal surfaces during CO oxidation under ultrahigh vacuum conditions [4], or as spiral waves in intracellular calcium release [5]. Further examples include circulating waves of neuromuscular activity in cardiac muscle tissue [6], and cyclic adenosine monophosphate (cAMP) waves in aggregating social amoeba colonies like the slime mould *Dictyostelium discoideum* [7]. A variant of the BZ reaction that uses the photosensitive complex rutheniumbipyridyl as redox catalyst [8] is a good candidate for studying the effect of external stimulation on spiral wave dynamics experimentally. Under illumination the reduced catalytic complex becomes photochemically excited and releases the inhibitor of the reaction, bromide. Therefore the local excitation threshold of the medium depends on the applied light intensity. Roughly speaking, high (low) intensity corresponds to low (high) excitability of the medium [9].

Well-known responses of spiral waves to external periodic forcing at a fixed frequency are synchronization and resonance. Synchronization is associated with the existence of entrainment bands where the response is phase locked. Periodic forcing at a frequency close to the rotation frequency of the spiral leaves the shape of the tip trajectory almost unchanged but results in a linear drift of the spiral core [10–15]. For feedback-controlled forcing several control modes are feasible. Grill, Zykov, and Müller [16,17] used information about the activity level at a particular recording point to control the intensity of spatially uniform light pulses. At a fixed time delay after a certain threshold was reached at the detector, a light pulse of given amplitude and duration was applied to the medium. It was demonstrated that the resonant drift may result in a motion of the spiral core along a circular pathway around the position of the detector, which imposes a center of symmetry for the dynamics. The radius of this so-called resonance attractor depends on the properties of the medium and can be changed by varying the delay time of the feedback loop [18].

We have realized this feedback loop using an open gel reactor [19]. The catalyst is immobilized in a silicahydrogel layer of 0.5 mm thickness (active layer) prepared on a plate of frozen glass (diameter 63 mm). To protect the reaction zone from stirring effects in the chamber of the reactor, the active layer is covered by an inactive gel layer not loaded with the catalyst (buffer layer). Fresh premixed solutions of the BZ reagents malonic acid (MA), bromate, bromide, and sulfuric acid are pumped continuously through the reactor and diffuse through the buffer layer into the reaction zone. During the experiments the chamber volume (120 ml) is completely exchanged once per hour. Recipe parameters are fixed: From the initial concentrations $[\mathrm{NaBrO_3}]=0.21M$, $[\mathrm{H_2SO_4}]=0.31M$, $[\mathrm{MA}]=0.19M$, and $[\mathrm{NaBr}]=0.041M$ follow $[\mathrm{H^+}]=0.34M$, $[\mathrm{MA}]=0.14M$, and $[\mathrm{BrMA}]=0.03M$. The temperature inside the reactor is kept at a constant value of $25.0\pm0.5\,^\circ\mathrm{C}$.

The active layer is uniformly illuminated using a slide projector (Braun 250 W) which is directly connected to a computer-controlled power supply (Hewlett Packard HP, 6653A). The absorption maximum of the rutheniumbipyridyl complex used is 459 mm, with a full width at half maximum of about 50 nm. To get the intensity in the spectral range 410–510 nm the light source was calibrated using an optical bandpass filter (SchottBG25+GG420). Desired intensity signals are transferred via a fast network from the main image-processing computer. Pattern formation in the active layer is recorded by a charge-coupled device CCD camera (Sony AVC D7CE) and digitized with a frame grabber (Data Translation DT 3155) every 250 ms. The main control program is a compiled WINDOWS 32-bit application (VISUAL C++); it uses image-processing libraries (Impuls Vision) for pattern analysis and the HP-VEE Active X-Control for power supply. The experimental setup allows an arbitrary choice of the detector position during the experiment.

Spiral waves are created by breaking a wave front with the spot of intense light (spot diameter about 1 cm). At a background intensity of 0.17 $\mathrm{mW/cm^2}$ the spiral tip moves on a hypocycloidlike orbit resembling a flower with four lobes directed outward. The tip coordinates are recorded au-

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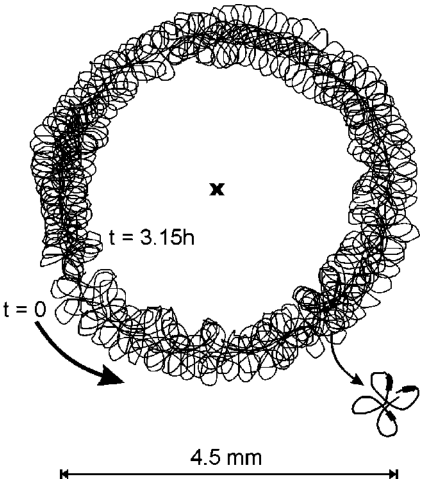

FIG. 1. Tip trajectory of a meandering spiral wave in a light-sensitive BZ medium obtained in an open gel reactor under feedback-controlled forcing. A light pulse (amplitude 0.29 mW/cm^2, duration 2 s, background illumination 0.17 mW/cm^2) is applied at every moment when the wave front has reached the detector, whose position is marked by a cross. In the lower right of the figure a part of the tip trajectory is selected. The shape of this tip path pattern is essentially the same as in the absence of forcing. Thick segments indicate the position of the tip during the application of the light pulse. The spiral wave is not in a synchronized state as the position of the thick segments changes from lobe to lobe.

tomatically. They are determined from the intersection of isoconcentration lines in consecutive images with 2.5 s spacing.

The width at the chosen intensity level is about one-quarter of the spiral pitch, which is $\lambda = 2.4 \pm 0.1$ mm. Far from the spiral core the medium is periodically excited with a frequency $f_{\mathrm{rot}} = \frac{1}{39}$ Hz. The propagation speed of the wave is 3.6 ± 0.1 mm/min.

Figure 1 illustrates the motion of the spiral tip under feedback-controlled pulsatory modulation. Each time the wave front crossed the measuring point, the applied light intensity was increased from 0.17 to 0.46 mW/cm^2 for 2 s. After some transients (not shown in the figure), the tip follows a circular pathway of radius R_S with the measuring point in the center. The tip motion is a superposition of compound rotation performed in the absence of forcing and circular resonant drift. The use of the open reactor is crucial as it takes 3 h 15 min to complete the whole pathway. The shape of the tip path pattern subjected to resonant drift is almost the same as the tip trajectory in the absence of forcing. There is no synchronization between the phase of the tip motion and the forcing.

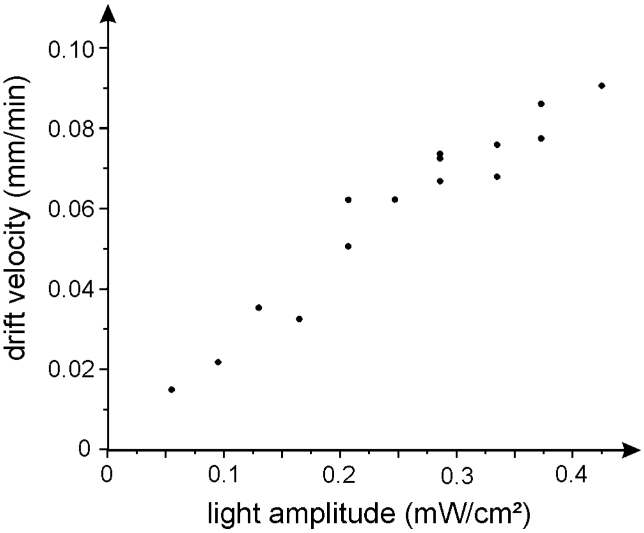

FIG. 3. Drift velocity of the spiral wave versus forcing amplitude. The data have been obtained from different branches of the resonance attractor.

Figure 2 displays the motion of the tip obtained in the same medium for three different initial positions of the detector with respect to the spiral tip. Tip trajectories, which in Fig. 1 are shown in detail, here for clarity are indicated by dark circular stripes [Fig. 2(a) corresponds to Fig. 1]. From the figure it is evident that there is a discrete set of possible radii R_S of stable branches of the resonance attractor with a constant spacing between neighboring values equal to the

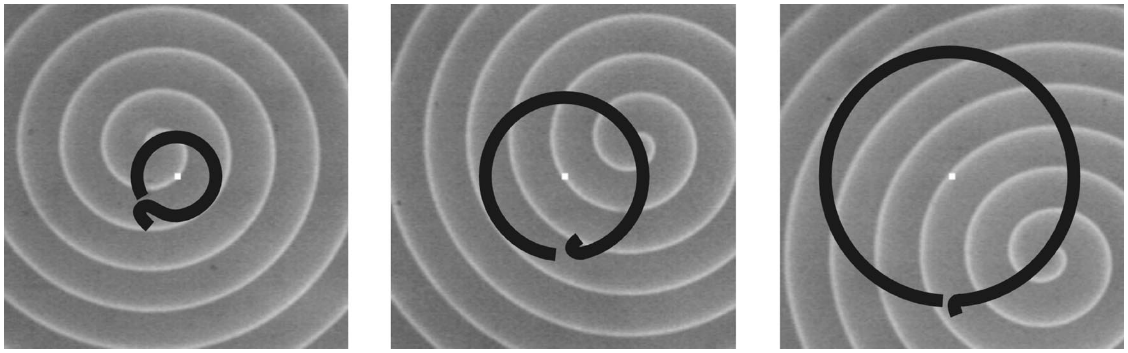

FIG. 2. Three stable branches of the resonance attractor of a meandering spiral wave observed in a light-sensitive BZ medium. Single branches are obtained for different positions of the detector marked by white dots. From the figure the λ spacing between neighboring stable radii R_S of the resonance attractor is evident. Forcing amplitude A and time for closing one pathway T are as follows: $A = 0.29$ mW/cm^2, $T = 3$ h 15 min (a); $A = 0.34$ mW/cm^2, T$= 6$ h50 min (b); and $A = 0.43$ mW/cm^2, $T = 7$ h50 min (c). Spiral rotation and motion along the branches of the resonance attractor are counterclockwise.

pitch λ of the spiral. This is in complete agreement with the results of a recent theoretical analysis by Karma and Zykov [18]. These authors reduced the dynamics of rigidly rotating spiral waves to a low-dimensional map describing the motion of the core center, and predicted an equidistant spectrum of fixed points R_S with a spacing of λ/2(λ) for the (stable) radii of the resonance attractor. In our experiments we observe the same behavior for meandering spiral waves. Note that, in order to obtain the pathway with the largest radius in Fig. 2, one has to maintain stationary nonequilibrium conditions over more than 700 rotation periods of the unperturbed spiral wave.

The forcing amplitude controls the drift rate, but not the size of the resonance attractor R_S. Compared to the wave speed the drift velocity is very slow. In accordance with results reported in [16,17], an increase of the forcing amplitude results in an increase of the angular velocity of the drift along the resonance attractor, leaving the radius unchanged. In our experiments we find a linear relation between modulation amplitude and drift velocity (Fig. 3). As the resonance is of first order, a linear relation is in agreement with the general mathematical theory as presented in [15]. For weakly excitable media a linear increase has been obtained in the framework of the so-called kinematical approach in the limit of small forcing amplitudes [20].

In our experiments feedback-controlled forcing turned out to be much more stable with respect to perturbations than external periodic forcing at a fixed frequency. We failed to obtain a stable resonant drift of spiral waves in the same system with external periodic forcing at the rotation frequency of the spiral. This may be important as the resonant drift of spiral waves has been discussed as a possible means to eliminate undesirable spiral wave sources in cardiac arythmia [21].

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