Period doubling in a periodically forced Belousov-Zhabotinsky reaction

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Using an open-flow reactor periodically perturbed with light, we observe subharmonic frequency locking of the oscillatory Belousov-Zhabotinsky chemical reaction at one-sixth the forcing frequency (6:1) over a region of the parameter space of forcing intensity and forcing frequency where the Farey sequence dictates we should observe one-third the forcing frequency (3:1). In this parameter region, the spatial pattern also changes from slowly moving traveling waves to standing waves with a smaller wavelength. Numerical simulations of the FitzHugh-Nagumo equations show qualitative agreement with the experimental observations and indicate that the oscillations in the experiment are a result of period doubling.

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I. INTRODUCTION

Periodic forcing of nonlinear oscillators produces a rich variety of dynamical responses, including frequency locking, quasiperiodic oscillations, period doubling, and chaos. These phenomena have been studied in the driven van der Pol [1–3] and other nonlinear differential equations [4,5], circle maps [6,7], analog circuits [8–10], cardiac tissue [11], and chemical systems [12]. In spatially extended oscillatory systems, periodic forcing can change the nature of the phase patterns from traveling waves or spiral waves in the unforced system to standing wave labyrinths [13,14] or multiphase spirals [15].

In this paper we present experimental observations of period-doubled chemical patterns for a light-forced Belousov-Zhabotinsky (BZ) reaction in resonance with the forcing. In earlier works, a sequence of subharmonic resonances was observed [16] that correspond to the Arnold resonance tongues of a single forced oscillator [17,18]. The Arnold tongues are arranged according to the Farey sequence, where the relation of the forcing frequency f_f to the response frequency f is in the ratio $m:n$ [19]. In each of the Arnold tongues, the temporal resonance affects the spatial pattern [20,21] and in addition adjusts the resonance regions in the forcing amplitude and frequency parameter plane [22].

We study a parameter range within the 3:1 resonance tongue of our experimental system. The pattern response is in resonance with the forcing frequency f_f and responds primarily at a frequency of $f_f/3$. Inside this region our observations reveal a response with large spectral power at $f_f/6$ where we expect to observe an $f_f/3$ response. We analyze the phase space trajectories of these patterns to distinguish between period-doubled 6:2 oscillations in a 3:1 tongue and the alternative frequency locking in the 6:1 tongue. We make a detailed comparison to a forced FitzHugh-Nagumo reaction-diffusion model, which clearly demonstrates period doubling in the 3:1 tongue.

In spatially extended systems, period doubling has been studied in parametrically forced coupled pendula [23] and in the Belousov-Zhabotinsky reaction [24–28]. The previous studies of period doubling in the BZ reaction have focused on period doubling of spiral waves including localized periodic forcing of the spiral wave tip [29]. Our approach is to explore the dynamics and pattern formation of periodic forcing in resonance with the spatially uniform BZ oscillation frequency.

II. EXPERIMENTAL METHODS

The Belousov-Zhabotinsky reaction is studied using an open-flow reactor system that allows for continuous supply of the chemical reagents [16,20,30]. The reaction takes place in a thin porous Vycor glass membrane, 0.4 mm thick and 22 mm in diameter, sandwiched between two chemical reservoirs. The reagents diffuse homogeneously from the continuously stirred reservoirs into the glass through its two faces. Each of the two 8.3 ml volume reservoirs (reservoirs A and B) are continuously refreshed at a flow rate of 20 ml per hour. They contain 0.8M sulfuric acid (A,B); 0.184M potassium bromate (A,B); 0.001M tris(2,2′- bipyridyl)dichlororuthenium(II)hexahydrate (A); 0.32M malonic acid (B), and 0.30M sodium bromide (B). Under these conditions, the reaction is oscillatory, and we observe rotating spiral waves of Ru(II) concentration in the membrane.

We image the spiral waves by passing spatially homogeneous low-intensity light through the membrane, and measure the relative intensity of the transmitted light using a charge-coupled device camera (~13 pixels/mm) bandpass filtered from 420 to 500 nm. Regions of the glass membrane that contain Ru(II) absorb light at 451 nm, so regions of high intensity have a lower concentration of Ru(II). Since the typical pattern wavelength is ⩾0.5 mm and the membrane is 0.4 mm thick, the pattern is quasi-two-dimensional.

The ruthenium catalyst of the BZ reaction is photosensitive in the visible range [31], and we apply forcing by shining a spatially uniform light on the membrane [30]. The light is applied using a commercial video projector (Sanyo PLC-750M) and a condenser lens. The output of the video projector is computer controlled using a video card with a refresh rate faster than 0.1 s. Using the computer controller, we modulate the spatially uniform light periodically in time with a square wave between low and high intensity. Both the light

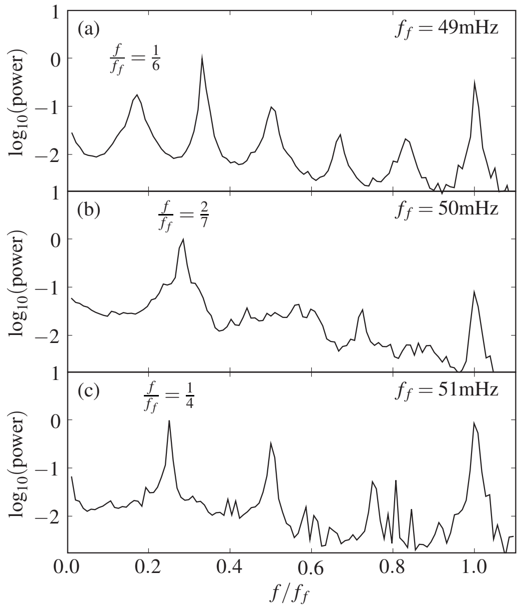

FIG. 1. (a) Subharmonic response at one-sixth the forcing frequency $f/f_f=1/6$ for periodic forcing at $f_f=49$ mHz. (b) At 50 mHz the dominant response is at $f/f_f=2/7$ and (c) at 51 mHz it is at $f/f_f=1/4$. The responses in (b) and (c) are consistent with each other but contrary to (a) in the ordering of the Arnold tongues. Since the frequency in (a) is half the expected $f/f_f=1/3$, this indicates a period-doubled response. The large peak at $f/f_f=1$ is from the imaging of the forcing light and is an artifact of the experimental setup, and not a large response from the chemical system.

intensity and frequency can be adjusted to study the effect of periodic forcing on pattern formation in the BZ reaction.

III. EXPERIMENTAL RESULTS

A. Power spectra

Without light forcing, the BZ reaction oscillates with a natural frequency f_0 that depends on the chemical conditions [32]. With light forcing of sufficient amplitude and with a frequency f_f near an integer ratio of the natural frequency, $f_f \approx (m/n)f_0$, the system responds by adjusting the response oscillation frequency $f=(n/m)f_f$ even though $f \neq f_0$. Regions of subharmonic frequency locking, or Arnold tongues, can be mapped out by varying the forcing frequency and amplitude [33]. We label the resonance tongues according to the ratio $m:n$. In the following, we will be concerned with the 3:1 tongue ($f_f \approx 3f_0$) and the nearby resonances.

The response to the forcing frequency is found by examining the power spectra of the resulting pattern. We apply the discrete Fourier transform in time at each pixel $p(x,y)$ in the pattern and then plot the average of the power spectral density vs frequency. Peaks in the subharmonic structure of the power spectral density indicate the resonant response.

When forcing with a frequency $f_f \approx 3f_0 = 49$ mHz, we observe a response with large spectral power at $f/f_f=1/6$ as shown in Fig. 1(a). In this range of forcing parameters, we expect $f=f_f/3$ or perhaps some nearby ratio like 2/7 or 1/4. The ratio f/f_f of response frequency to forcing frequency should decrease when the forcing frequency is increased, following the Farey sequence [16,34]. But at slightly higher forcing frequency $f_f=50$ mHz we observe a peak in the response at $f/f_f=2/7$ [Fig. 1(b)]. This is larger than the response at $f_f=49$ mHz and contrary to the ordering of the Arnold tongues. At $f_f=51$ mHz, we observe a response at $f/f_f=1/4$, which is lower than at $f_f=50$ mHz (and follows the correct ordering) but still higher than at $f_f=49$ mHz [Fig. 1(c)].

One possible explanation for the response at $f/f_f=1/6$ when we expect 1/3 is period doubling. We test for period doubling in the experiment by starting with a 3:1 resonant response ($f/f_f=1/3$) for parameters near those of the 1/6 response. We slowly increase the forcing strength while maintaining a constant forcing frequency. The initial forcing strength and frequency are chosen such that the response is 3:1 resonant and the spatial pattern consists of large uniformly oscillating domains at one of the three stable phases. Figure 2(a) shows the the power spectrum with a large peak at $f/f_f=1/3$ and harmonics at integer multiples of that frequency. When the forcing strength is increased (keeping the frequency the same), a new peak forms at $f/f_f=1/6$ [Fig. 2(b)] at nearly the same power as at $f/f_f=1/3$ indicating a 6:2 period-doubled response.

B. Phase space trajectories

In addition to the power spectra, we reconstruct a two-dimensional phase space from our intensity vs time measurements using a time delay coordinate method. Similar methods have been demonstrated in numerical systems [35] and BZ systems where a single variable is measured [36]. For a fixed point in the pattern, $p_0=w(x,y)$, we plot $p_0(t)$ vs $p_0(t-\tau)$ with τ chosen to capture the structure of the orbit in phase space. The point p_0 is a representative point from the interior of the homogeneously oscillating 3:1 domains. For the data in Fig. 2, $\tau=10 \approx 0.49/f_f$. As the forcing strength is increased, the single-orbit trajectory shown in in Fig. 2(c) is twisted into a double-orbit trajectory shown in Fig. 2(d). The signature of period doubling in this phase space is two distinct loops. Instead of the same oscillation every three forcing cycles, there are two distinct oscillations, one small and one large loop, every six cycles. This is distinctly different from the response for a forced system when $f_f \approx 6f_0$, which also has significant spectral power at $f_f/6$ but makes only one oscillation in six forcing cycles.

In addition to the temporal response, spatial pattern formation also gives us another piece of evidence for period doubling. In order to clearly see the patterns, we process the data and filter to keep only the frequencies with significant power. Let $p(x,y,t)$ represent the light signal recorded by the camera in the reactor system. We Fourier transform the signal, as above, to get the power spectrum, use a bandpass filter to select out a set of frequencies to analyze, and then inverse-transform the filter signal. If we filter the data only near $f/f_f=1/3$ and 1/6, the result is a representation of the data as

$$p(x,y,t) = A_6(x,y,T)e^{i\omega_f t/6} + A_3(x,y,T)e^{i\omega_f t/3} + \text{c.c.}, \quad (1)$$

where $\omega_f=2\pi f_f$, and $A_6(x,y,T)$ and $A_3(x,y,T)$ represent the complex amplitude of the signal at $f/f_f=1/6$ and 1/3. The

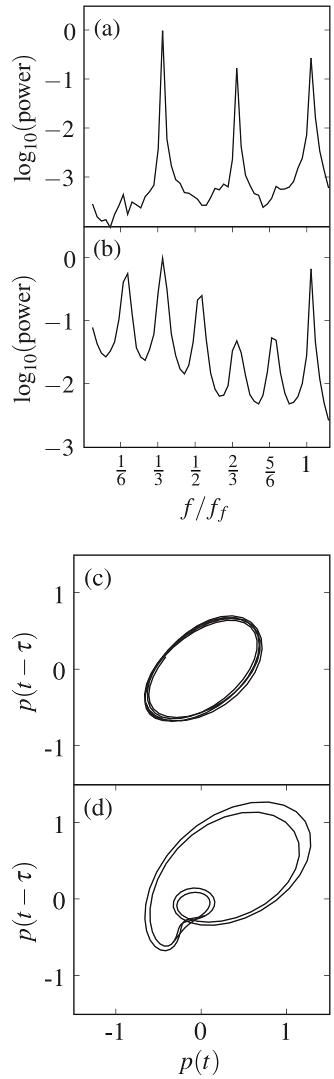

FIG. 2. (a) Power spectrum of a 3:1 resonant response. (b) Power spectrum of a 6:2 resonant period-doubled response. (c) Phase space trajectory in the 3:1 response makes a single loop every three forcing cycles. (d) Phase space trajectory in the 6:2 period-doubled response makes two distinct loops for every six forcing cycles. Chemical parameters are given in Sec. II; forcing frequency 50 mHz, forcing strength in (a) and (c) 33.6 W/m^2, in (b) and (d) 37.2 W/m^2.

amplitude and phase of A_6 and A_3 vary over space and possibly in time on a time scale T much longer than the forcing period $1/f_f$.

The phase of the 3:1 pattern response, arg(A_3), with no period doubling is either three-phase spiral waves or slowly moving irregular-shaped domains of three phases [21]. In the period-doubled 3:1 response, the same type of irregular-shaped domains are found at $f/f_f=1/3$, but at $f/f_f=1/6$ the pattern phase, arg(A_6), is a standing wave consisting of six phases as shown in Fig. 3. This is in sharp contrast with the 6:1 pattern response of six-phase spiral waves reported previously [21].

In addition to the difference in the spatial phase organization of the pattern, the A_6 response observed through a period doubling in the 3:1 tongue has significantly different phase

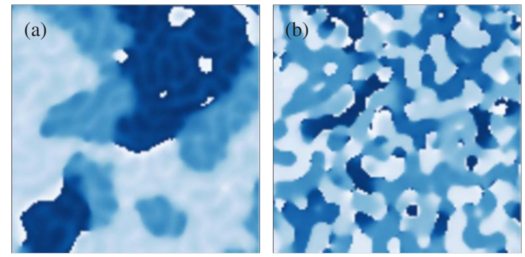

FIG. 3. (Color online) Patterns formed in a 128×128 pixel region of the BZ reactor with parameters corresponding to a 3:1 response to the forcing. (a) The pattern at $f/f_f=1/3$ is irregularly shaped domains. (b) The pattern at $f/f_f=1/6$ is shorter-wavelength standing waves. The higher harmonics have linear combinations of these two basic patterns. Chemical parameters are given in Sec. II; forcing frequency 49 mHz, forcing strength 37.8 W/m^2.

structure from the response at higher-frequency forcing. By plotting the phase arg(A_6) in the complex plane two distinct shapes are revealed as shown in Fig. 4. Both cases have a sixfold symmetry corresponding to six stable uniform phases, which are found at the points farthest from the center. The interfaces connecting these phases in space to make a pattern are very different. In the 6:1 tongue, the states are connected with traveling fronts that shift the phase by $\pi/3$ and do not go through the origin. The period-doubled pattern phases are connected by fronts that go through the origin (standing waves) where the complex amplitude goes to zero. In this case, the phase difference between neighboring spatial regions is π or $2\pi/3$.

The higher harmonics not included in the expansion in Eq. (1) do not contain any additional pattern formation features for the period-doubled system. We have found that there are two independent modes, but not more. For example, the phase of the A_2 signal (the response at $f/f_f=1/2$) is, in fact, just the sum of the phases at A_6 and A_3, the previous two,

$$\arg(A_2) = \arg(A_6) + \arg(A_3). \qquad (2)$$

IV. THE PERIODICALLY FORCED FITZHUGH-NAGUMO MODEL

We use a version of the FitzHugh-Nagumo (FHN) reaction-diffusion equations as a model for a periodically

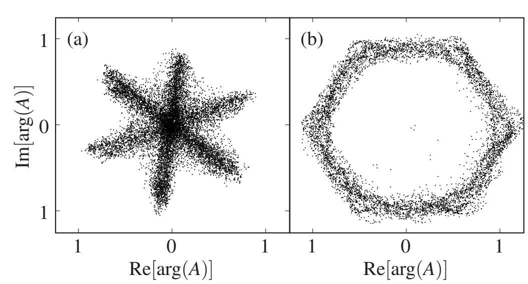

FIG. 4. Phase of the pattern for an $f/f_f=1/6$ response in the 3:1 tongue and 6:1 tongue. (a) The period-doubled standing-wave pattern in the 3:1 tongue. (b) The spiral wave in the 6:1 tongue. The two different cases are clearly topologically distinct. Chemical parameters are given in Sec. II; forcing parameters 49 mHz and 37.8 W/m^2 in (a) and 100 mHz and 60.29 W/m^2 in (b).

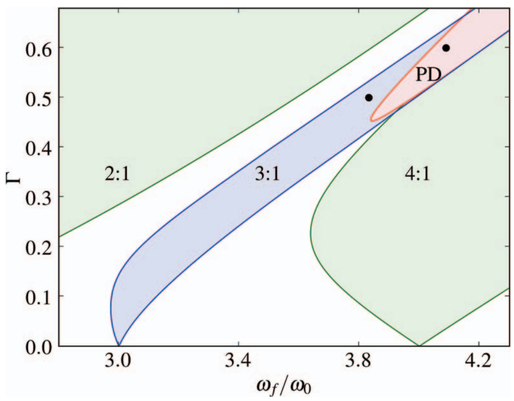

FIG. 5. (Color online) Resonance boundaries for the 2:1, 3:1, and 4:1 tongues for the periodically forced FHN equations (3). The horizontal axis spans the ratio of the forcing frequency ω_f to the natural frequency, $\omega_0 \approx 0.214$, of the unforced system. Within the tongue boundaries, uniform solutions may lock at $m:n$ ratios of the forcing. In the upper part of the 3:1 tongue, there is a region of period doubling (labeled PD). Further period doublings or chaotic oscillations may occur for higher forcing in the 3:1 tongue and in the upper regions of the 2:1 and 4:1 tongues. The parameters in the FHN equations are $\epsilon=0.1$, $a_1=0.5$, $a_0=0.05$, and $\delta=0.1$.

forced oscillatory system. We modify the FHN model to include time-periodic forcing,

$$u_t = u - u^3 - v + \nabla^2 u, \tag{3a}$$

$$v_t = \epsilon(u - a_1 v - a_0) + \Gamma \sin(\omega_f t) v + \delta \nabla^2 v. \tag{3b}$$

The fields $u(x,y)$ and $v(x,y)$ represent the concentrations of activator and inhibitor types of chemical reagents. The periodic forcing is provided by the sinusoidal term with amplitude Γ and frequency ω_f. The parameter ϵ is the ratio of the characteristic time scales of u and v, and δ is the ratio of the diffusion rates of u and v. In the following we use a one-dimensional version of Eqs. (3) where the fields are functions of only one space variable x.

With no forcing, $\Gamma=0$, Eq. (3) has a spatially uniform solution $u=u_0$, $v=v_0$ with a Hopf bifurcation to uniform oscillations at the critical point $\epsilon=\epsilon_H$. The Hopf point for the symmetric model ($a_0=0$) is $\epsilon_H=1/a_1$ and the natural oscillation frequency is $\omega_0=\sqrt{1/a_1-1}$. In addition to the uniform oscillations, traveling waves including spiral waves exist below the Hopf bifurcation $\epsilon<\epsilon_H$.

When forced with sufficient amplitude Γ and with frequencies near rational multiples of the natural frequency, $\omega_f/\omega_0=m/n$, the system may lock to the forcing frequency. The locking regions form resonance tongues (Arnold tongues) in the ω_f-Γ parameter plane. Inside the resonance tongues, the system adjusts its oscillating frequency to a specific rational multiple of ω_f. For example, for forcing near three times the natural frequency $\omega_f \approx 3\omega_0$, the system adjusts to $\omega_f/3$ and is in the 3:1 resonance tongue. Figure 5 shows the resonance boundaries for the 3:1 resonance tongue with the nearby 2:1 and 4:1 tongues.

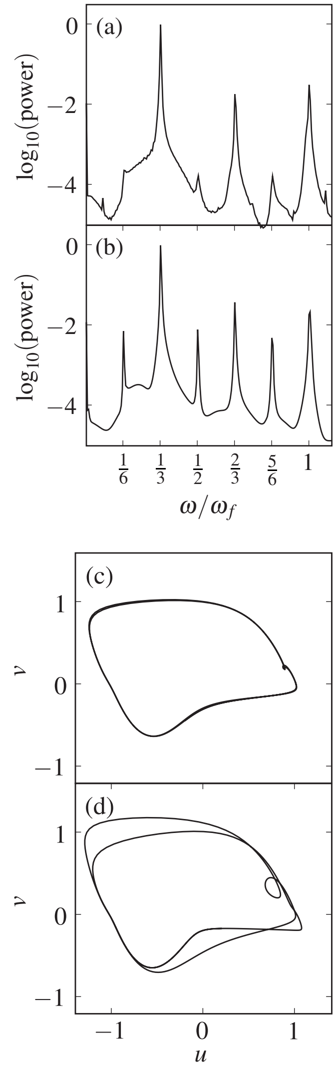

FIG. 6. Power spectra and corresponding phase trajectories for the 3:1 response and period-doubled response in the forced FHN equations (3). (a) The power spectrum for the 3:1 response shows a primary peak at $\omega/\omega_f=1/3$. (b) The power spectrum for the 3:1 period-doubled response shows a frequency response at $\omega/\omega_f=1/6$ in addition to the higher harmonics. (c) The orbit of the 3:1 response in the u-v plane. (d) The orbit of the period-doubled response in the 3:1 tongue. The parameters are the same as in Fig. 5 with the parametric forcing amplitude and frequency indicated by the solid circles. (a),(c) $\Gamma=0.5$, $\omega_f=0.82$. (b),(d) $\Gamma=0.6$, $\omega_f=0.875$.

For large enough forcing amplitude inside the 3:1 tongue there is a region where the period of the 3:1 resonant orbit doubles. In this region of period doubling, the effective frequency of the system response is halved, which is easily observable in the power spectra. Figure 6 shows the power spectra and orbits for the 3:1 response and the 3:1 period-doubled response in Eqs. (3). The period-doubled response shows the lowest-frequency peak in the power spectra at $\omega_f/6$ (and with higher harmonics) as opposed to $\omega_f/3$ for the 3:1 response. The orbits in the u-v phase plane also show a transition from a single loop to a double loop over the same time period of three forcing cycles. These observations are similar to those in the BZ reaction.

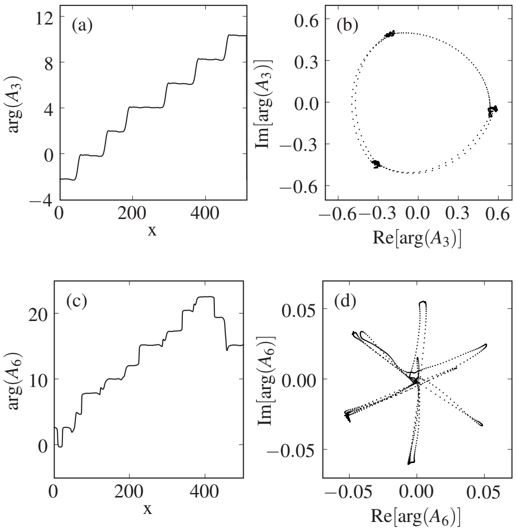

FIG. 7. Phase patterns for the 3:1 period-doubled FHN equations (3). (a) The response at $\omega_f/3$ is a traveling wave consisting of three phases. (b) The complex phase plane representation of the $\omega_f/3$ response shows that the fronts shift the phase by $2\pi/3$ and do not go through the origin. (c) The response at $\omega_f/6$ is a standing wave consisting of six phases. (d) The complex phase plane representation of the $\omega_f/6$ response shows that the fronts always cross through zero (the node of the standing wave) and can shift the phase by either π or $2\pi/3$.

The typical patterns in the 3:1 tongue of the forced FHN model (3) are traveling waves between the three stable phases. In two-dimensional systems these waves may form rotating spiral patterns if the system size is large enough to accommodate the spiral [21]. In the period-doubled parameter region the traveling pattern is accompanied by a standing-wave stationary pattern at $\omega=\omega_f/6$ as in the BZ system. Figure 7 shows the pattern response at $\omega_f/3$ and $\omega_f/6$ using the same analysis method as for the experimental data [see Eq. (1)]. The pattern is shown both as a function of space and in the complex plane of the phase.

For the FHN solutions in Fig. 7 the phase in space as shown as a cumulative phase to demonstrate that the phase shifts for the fronts are always $2\pi/3$ for the $\omega_f/3$ response and can be either $2\pi/3$ or π through the possible six phases in the $\omega_f/6$ response. The complex phase plane shows that the fronts between the six phases always traverse through zero (the zero is a node position), indicating a standing-wave pattern as in the BZ system.

V. CONCLUSION

We have demonstrated period-doubling dynamics in both the BZ chemical reaction and a periodically forced version of the FHN equations. The period doubling is found in a parameter range inside the 3:1 resonance tongue and leads to a subharmonic signal at $\omega_f/6$ where we would expect the lowest-frequency signal to be $\omega_f/3$. Examination of the oscillation orbits and phase structure of the pattern formation gives further evidence that the phenomenon is a result of period doubling.

It is not possible in the experiment to rule out an overlap of 2:1 and 3:1 resonance regimes as an alternative explanation for the low-frequency response we see inside the 3:1 resonance tongue. The interaction of the 2:1 and 3:1 modes may lead to a subharmonic signal at $f_f/6$. This explanation is ruled out in the FHN model where the tongue boundaries are well defined and the period-doubling regime falls entirely in the 3:1 tongue where there is no overlapping with the 2:1 tongue. The similarities between the experiment and the simulation we have presented here favor period doubling as the explanation for our experimental observations.

Period doubling should occur in other tongues depending on the exact nonlinear dynamics of the chemical or model system. We have also observed possible period doubling in the 2:1 resonance tongue, but have not yet verified that this is the case. In addition there are likely further period doublings inside the region we have found and, as in other systems, there could be an entire period-doubling cascade to chaos [3].

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