Transition from Simple Rotating Chemical Spirals to Meandering and Traveling Spirals

Ge Li,* Qi Ouyang, Valery Petrov, and Harry L. Swinney

Center for Nonlinear Dynamics and Department of Physics, The University of Texas at Austin, Austin, Texas 78712

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Experiments on the Belousov-Zhabotinksy reaction unfold the bifurcation from simple (temporally periodic) rotating spirals to meandering (quasiperiodic) spirals in the neighborhood of a codimension-2 point. There are two types of meandering spirals, inward-petal (epicycloid) spirals and outward-petal (hypocycloid) spirals. These two types of meandering regimes are separated in the phase diagram by a line of traveling spirals that terminates at the codimension-2 point. The observations are in good accord with theory. [S0031-9007(96)01014-9]

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Rotating spiral waves are ubiquitous in systems ranging from excitable reaction-diffusion media [1,2] to aggregating slime-mold cells [3] to cardiac muscle tissue [4]. Winfree discovered that under certain conditions a spiral tip *meanders* rather than follows a periodic circular orbit [5]. Meandering spirals have been subsequently extensively studied experimentally [6–9] and theoretically [10–16]. Experiments [8,9] and theoretical analyses [12,13,16] have shown that the meandering is often not an erratic motion; rather, the spiral tip moves in epicycloid-like [17] orbits (flowerlike orbits with inward petals) or hypocycloidlike orbits [17] (with outward petals) that are *B* quasiperiodic in time [8]. Meandering is of interest in part because of its predicted relation to defect mediated turbulence [18–20]. It may also provide a clue to the cause of cardiac arrythmias, which can lead to ventricular fibrillation [21].

There have been few definitive experimental results on meandering other than the observation that the onset of meandering is a periodic-quasiperiodic transition [8,22]. We present here experiments on the Belousov-Zhabotinsky (BZ) reaction that reveal, as predicted by Barkley [16], an unfolding of the bifurcation to meandering about a codimension-2 point that is the terminus of a line of traveling spirals.

Figure 1 shows the orbit of a spiral tip for the two types of meandering motion [23]: (a) an outward-petal meandering spiral and (b) an inward-petal meandering spiral. We take the spiral tip to be the point with maximum local curvature on the wave front. Hypocycloid motion with outward petals is illustrated in Fig. 1(c), where the primary circle (radius r_1) orbits the secondary circle (radius r_2) in one direction with frequency f_2 and spins about its center in the opposite direction with frequency f_1; epicycloid motion with both rotations in the same sense and inward petals is illustrated in Fig. 1(d).

Figure 2 illustrates all four types of spiral motion that we have observed. Figure 2(a) is a simple periodic rotating spiral, which becomes unstable as a parameter is varied. Depending on the control parameter, the system then chooses outward-petal meandering, as in Fig. 2(b), or inward-petal meandering, as in Fig. 2(d). The tip of a meandering spiral emits waves that are compressed in front of the tip and dilated behind the tip. This produces superspirals, as can be seen in Fig. 2(b), which has a retrograde superspiral, and in Fig. 2(d), which has a prograde superspiral. At the transition from outward-petal to inward-petal spirals, the spiral tip travels in a straight line; see Fig. 2(c).

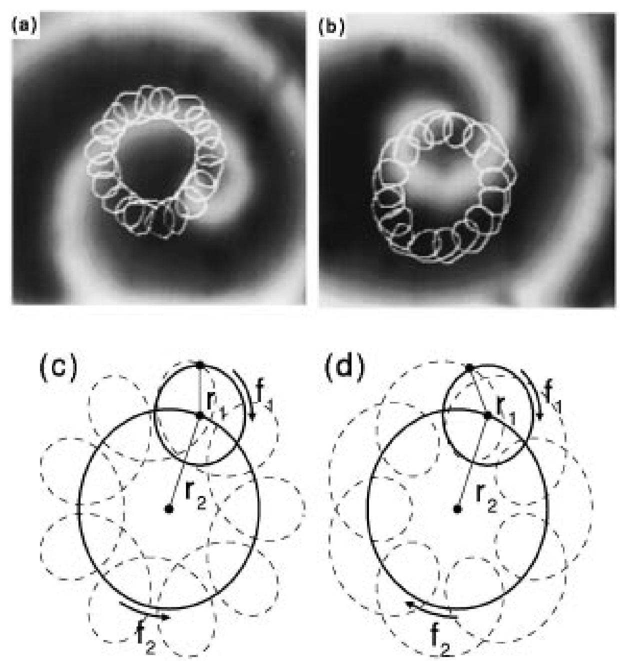

FIG. 1. Meandering spiral with (a) outward and (b) inward petals. The white lines in the images show the trajectories of spiral tip. (c) and (d) illustrate, respectively, a hypocycloid and an epicycloid, analogous to the motion in (a) and (b). The only parameter different in (a) and (b) is the concentration of sulfuric acid in reservoir B: (a) $0.46M$, (b) $0.40M$. The other control parameter in the present experiments is the concentration of malonic acid in reservoir B, which is fixed in this figure and Figs. 2 and 3 at $0.8M$ but varied in Fig. 4. Other conditions are fixed in our experiments at the values given in Ref. [23]. The pictures in (a) and (b) are 1.7×1.7 mm^2.

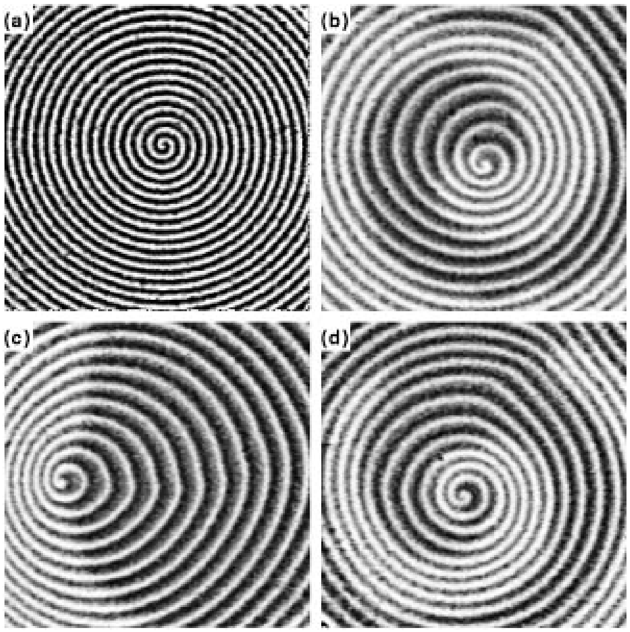

FIG. 2. (a) A simple rotating spiral with period 8.1 s and wavelength 0.42 mm, (b) a meandering spiral with outward petals, (c) a traveling spiral, and (d) a meandering spiral with inward petals. In (b)–(d) the superstructure reflecting the Doppler effect is visualized by the following procedure: the image of a superstructure is first extracted from the original picture using nonlinear image processing, then the extracted image is superimposed on the original picture. The sulfuric acid concentration in reservoir B is the control parameter that is varied: (a) $0.67M$, (b) $0.46M$ [as in Fig. 1(a)], (c) $0.44M$, (d) $0.40M$ [as in Fig. 1(b)]. The other parameters are fixed at the value given in Fig. 1. Each image is 12.7×12.7 mm^2.

Our experiments are conducted in the ferroin-catalyzed BZ reaction in an open spatial reactor, as described previously [24]. The reaction occurs in a thin porous glass disk, 0.4 mm thick and 25.4 mm in diameter (Vycor glass, Corning). The Vycor disk prevents advection in the reaction medium, thus allowing only reaction and diffusion processes. Each surface of the disk is in contact with a continuously fed stirred reservoir where the reactants are maintained homogeneous and far from thermodynamic equilibrium. The two reservoirs are labeled differently (reservoir A and reservoir B) because they have different reagent concentrations. The control parameters in the experiments are $[H_2SO_4]_0^B$ and $[CH_2(COOH)_2]_0^B$. The catalyst concentration in the Vycor increases in going from reservoir B to reservoir A, while the malonic acid concentration decreases in going from reservoir B to reservoir A. The chemical spatial patterns form only in a thin layer in the Vycor where the oppositely directed chemical gradients cross. Thus the thickness of the pattern forming layer is small compared to the wavelength of the spirals. We assume that the observed patterns are quasi-two-dimensional [25].

We study the behavior of single spirals, which can be moved in position using red laser light ($\lambda = 632.8$ nm). If there are initially several spirals, laser light is applied to move one tip to the center and the others to the disk edge. If there is initially no spiral tip, a counterrotating pair is created by focusing the laser light on a point in the medium; then one tip is removed. Laser light is also used occasionally to change the direction of a spiral tip; otherwise a spiral like the one in Fig. 2(c) would hit the edge of the reactor and die.

We have examined the spiral motion as a function of decreasing sulfuric acid concentration in reservoir B [$(0.70-0.25)M$] with other reagent concentrations held fixed; see Fig. 3. Meandering spirals exist only in the window $0.67 > [H_2SO_4]_0^B > 0.26M$. Outside this window the spiral tips follow circular orbits; in this case a spiral viewed in a corotating reference frame is time independent [13]. As $[H_2SO_4]_0^B$ is decreased (increased) across a critical value ($0.67M$ and $0.26M$, respectively), simple spirals become unstable and the system undergoes a transition to outward (inward) petal meandering spirals. The meandering motion is quasiperiodic; the ratio of the two frequencies changes continuously with control parameters—there is no locking of the two frequencies in an integer ratio. The radius of the second cycle r_2 grows continuously as $[H_2SO_4]_0^B$ approaches a critical concentration ($0.44M$) from either above or below, as Fig. 3 illustrates. This critical concentration marks the transition from outward petal to inward petal meandering spirals as the concentration is decreased.

A normal form analysis by Barkley [16] shows that meandering behavior can be understood by studying the region around a codimension-2 point that is the terminus of the line dividing the domain with meandering into two regions, one with inward petals and the other with outward petals. We achieve this unfolding of the bifurcation using

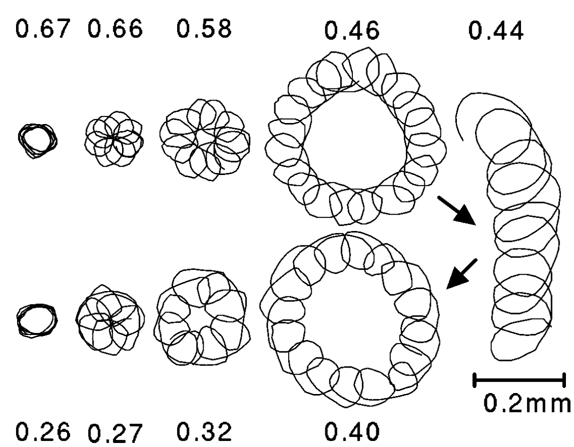

FIG. 3. Spiral tip orbits observed as a function of the sulfuric acid concentration in reservoir B (the numbers are molar concentrations): $0.67M$, a circular orbit; $(0.66-0.46)M$, hypocycloidlike orbits with outward petals; $0.44M$, a traveling tip; $(0.40-0.27)M$, epicycloidlike orbits with inward petals; $0.26M$, a circular orbit. The other parameters are fixed at the values given in Fig. 1. With decreasing $[H_2SO_4]_0^B$ in the range $(0.7M, 0.25M)$, the primary period of rotation increases from 8.0 to 12.0 s, and the average wavelength, determined by Fourier transform, increases from 0.40 to 0.59 mm.

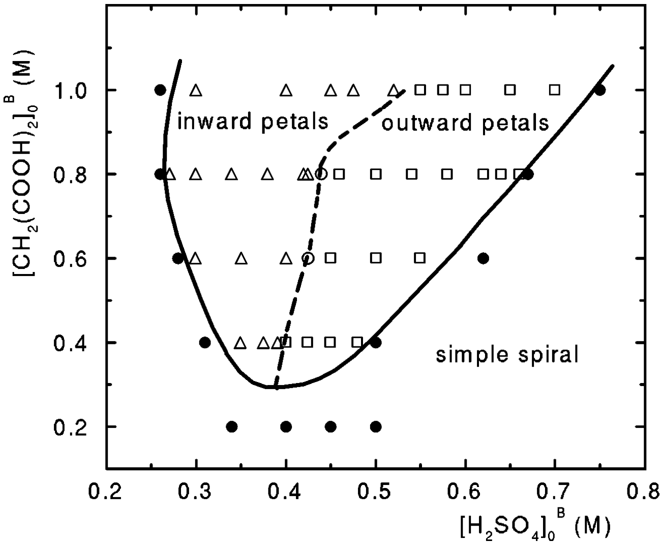

FIG. 4. Dynamics of spirals as a function of $[H_2SO_4]_0^B$ and $[CH_2(COOH)_2]_0^B$ (with other conditions as in Fig. 1). The solid line marks the transition from simple spirals (●) to meandering spirals with inward (△) and outward (□) petals. Traveling spirals (○) exist along the dashed line that separates the two types of meandering spirals.

$[CH_2(COOH)_2]_0^B$ and $[H_2SO_4]_0^B$ as control parameters, as Fig. 4 illustrates. Our phase diagram is qualitatively the same as that obtained by Barkley.

Studies of models show that the onset of meandering is a supercritical Hopf bifurcation [6,11–13,16]. We have examined both of the transitions from simple to meandering spirals, and our results are presented in Fig. 5(a), where r_1 is the average radius of a petal and $r_2 = D/2 - r_1$ (with D the outer diameter of the orbit).

The observations for both transitions agree with the prediction that the ratio of radii squared should increase linearly with control parameter distance from the transition. Far beyond each of the Hopf bifurcations the square of the radius ratio increases faster than linearly. The secondary radius diverges, yielding a traveling spiral [cf. Fig. 2(c)] as a critical control parameter value ($[H_2SO_4]_0^B = 0.44M$) is approached.

The normal form analysis by Barkley [16] predicts that, as the system approaches the line corresponding to a traveling spiral, the flower sizes r_2 diverge as $(\omega_1^2 - \omega_2^2)^{-1}$, where ω_1 is the primary frequency of a spiral and ω_2 is the frequency of spiral modulation in the rotating frame [16]. Our frequencies f_1 and f_2 correspond, respectively, to ω_1 and $|\omega_1 - \omega_2|$ in the notation of Barkley. Thus we have

$$r_2 \propto (\omega_1^2 - \omega_2^2)^{-1} = (f_1 f_2)^{-1}\left(1 + \frac{\omega_2}{\omega_1}\right)^{-1}. \quad (1)$$

Near the transition to traveling spirals, we have $\omega_1 \sim \omega_2$, so that

$$r_2 \propto (f_1 f_2)^{-1}. \quad (2)$$

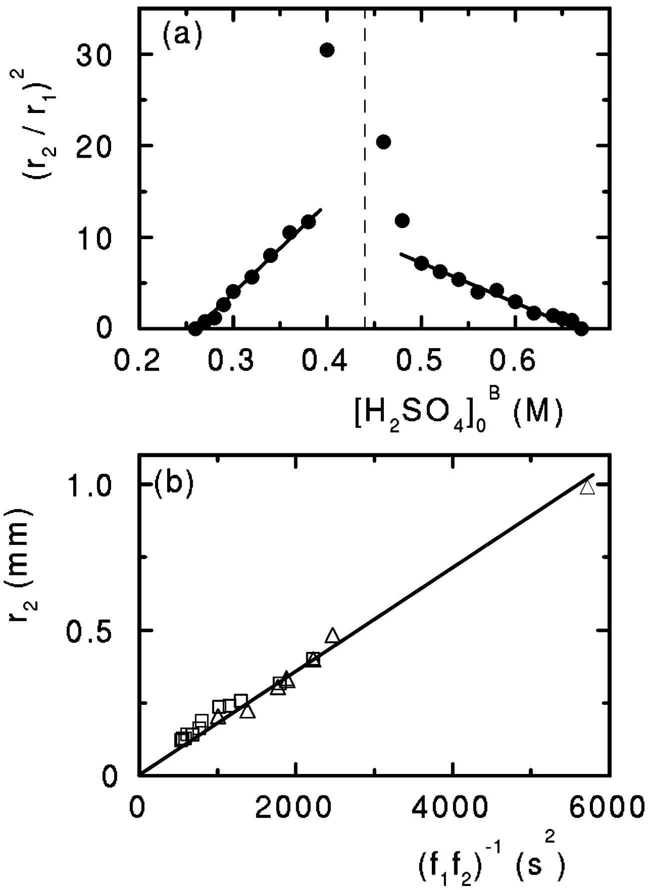

FIG. 5. (a) Evidence that both transitions from simple to meandering spirals are Hopf bifurcations: $(r_2/r_1)^2$ increases linearly with the control parameter beyond each transition. The vertical dashed line indicates the transition between inward- and outward-petal spirals; the radius ratio diverges as this line is approached. (b) Comparison of the measured r_2 with theory (solid line) as a function of $(f_1 f_2)^{-1}$ for both inward-petal spirals (△) and outward-petal spirals (□). Parameters other than $[H_2SO_4]_0^B$ are the same as in Fig. 1.

We determine f_1 by measuring the average frequency of spiral waves far from the spiral center, and we obtain f_2 from the relation $f_1/f_2 = n$, where n is the number of petals of the flower. Figure 5(b) shows that the observations for both inward petal and outward petal meandering spirals are in good accord with the theoretical prediction.

In conclusion, we have observed all four types of periodic and quasiperiodic spiral patterns predicted in theoretical and numerical studies [6,7,9,11–13,15,16]. Our experiments show that the transition from a simple to a meandering spiral is a supercritical Hopf bifurcation. Further, the data provide strong support for Barkley's analysis [16] of the normal form near the codimension-2 point where the locus of traveling spirals meets the boundary of the meandering region in the phase diagram (cf. Fig. 4). Also, the observed growth of the flower size as a function of the two rotation frequencies is in accord with the prediction of the normal form analysis. Finally, we mention that, for control parameter values near the codimension-2 point ($[H_2SO_4]_0^B = 0.38M$, $[CH_2(COOH)_2]_0^B = 0.30M$), the tip motion appears somewhat erratic. Future experiments can examine this behavior to determine whether it is spatiotemporal chaos or due to external noise.

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*On leave from Chemistry Department, Tsinghua University, Beijing, China.

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