Spontaneous scroll ring creation and scroll instability in oscillatory medium with gradients

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(Received 10 May 2005; revised manuscript received 6 October 2005; published 12 December 2005)

Scroll waves in a quasi-three-dimensional reaction-diffusion medium with a gradient in the third dimension are studied by numerical simulations using the Fitzhugh-Nagumo model. Under a simple initial condition with only one straight filament, we show a spontaneous creation of twisted scroll rings surrounding the initial filament when the control parameter is increased across a threshold. We find that due to the presence of the gradient, the difference of oscillation frequencies in the third dimension is the underlying cause of the phenomenon. Further increase of the control parameter will lead to the spiral breakups, as a result of the interaction of filaments. Observations in the experiments conducted in the same type of medium with the Belousov-Zhabotinsiky reaction qualitatively agree with our simulation results.

DOI: 10.1103/PhysRevE.72.066207 PACS number(s): 05.45.Xt, 82.40.Ck, 47.32.Cc

I. INTRODUCTION

Spiral waves have been widely observed in diverse physical, chemical, and biological systems. The transition of regular spirals to spatiotemporal chaos has been the focus of research in the past few years. It still remains a challenging problem at present [1,2]. Remarkable achievements has been documented; the mechanisms of two-dimensional (2D) spirals instability are understood: it can be caused by the convective (absolute) Eckhaus instability [3,4] and Doppler instability [5]. As to 3D scroll waves, filament dynamics is studied [6,7,10] and linear stability of scroll waves is analyzed [8,9]. A mechanism of a typical 3D instability is proposed as "negative tension instability" or "Winfree turbulence" [7,10–12]. It is suggested that a regular scroll wave pattern can become unstable while its filaments "snake about" in the medium [12]. Since the complexity of scroll waves is dominated by the structure of filaments, these organizing centers have been envisaged and topologically analyzed [13]. Among them, however, only untwisted scroll rings usually created artificially as initial condition are substantially studied in numerical simulations of reaction-diffusion models [7,12]. A scroll ring with phase twists cannot exist solitarily according to "Exclusion Principle" [13], so that it is difficult to obtain these rings in experiments or simulations in the reaction-diffusion system.

In the meantime, the influence of gradient in the medium on the spirals or scrolls is also studied. In 2D systems, the primary effects are global drifting and competition between several spirals [14,15]. In 3D systems, it is proposed that a gradient brings phase twists around the vortex filament and makes the scroll easier to break, either by extending the region of a classical type of instability [16,17] or by introducing new possible mechanisms of breakup [18]. The breakup caused by frequency differences and phase twists however, has not been observed in numerical simulations and experiments in an inhomogeneous excitable medium. On the contrary, it is found and believed that a parameter gradient curls the straight-line filament, making it form a helix, so that it leads to "sproing instability" [19–21].

In this paper, we show that there exists a new type of spiral instability in a 3D reaction-diffusion medium under the influence of a gradient. In this case, although the given straight filament is steady and stable, different patterns of filaments can spontaneously appear in other regions of the medium, breaking the symmetry of the system and leading to spatiotemporal chaos. The destabilization mechanism differs from both typical 3D negative-tension instability and 2D Eckhaus or Doppler instability. By examining the numerical details such as local frequency, phase difference along a line, and distribution of defects, we reach a qualitative explanation of filament formation and the ultimate symmetry breaking. The origin of this type of instability is much like what is depicted in Ref. [18], but the process is much more complicated. Many details are shown in the following text, including periodic breakup, spontaneous scroll ring creation, and complex but symmetric scroll patterns. What is more, the scroll rings spontaneously appearing in the system is intrinsically twisted and punctured by a straight filament, so we obtained twisted scroll rings that Winfree envisaged. Some of our simulation results are supported by our experimental observations of the Belousov-Zhabotinsiky (BZ) reaction.

This paper is organized as follows: After introducing the mathematical model system in Sec. II, we report our observations in numerical simulations in Sec. III, where we show the details of the scroll ring and "hot dog" formations and the spiral breakup scenario. Experimental observations that support our numerical results are also presented in this section. In Sec. IV, we give an explanation on the mechanism of the filament creation, followed by a brief conclusion.

II. MODEL

In numeric simulation we use a classical Fitzhugh-Nagumo model, which consists of two chemical species u

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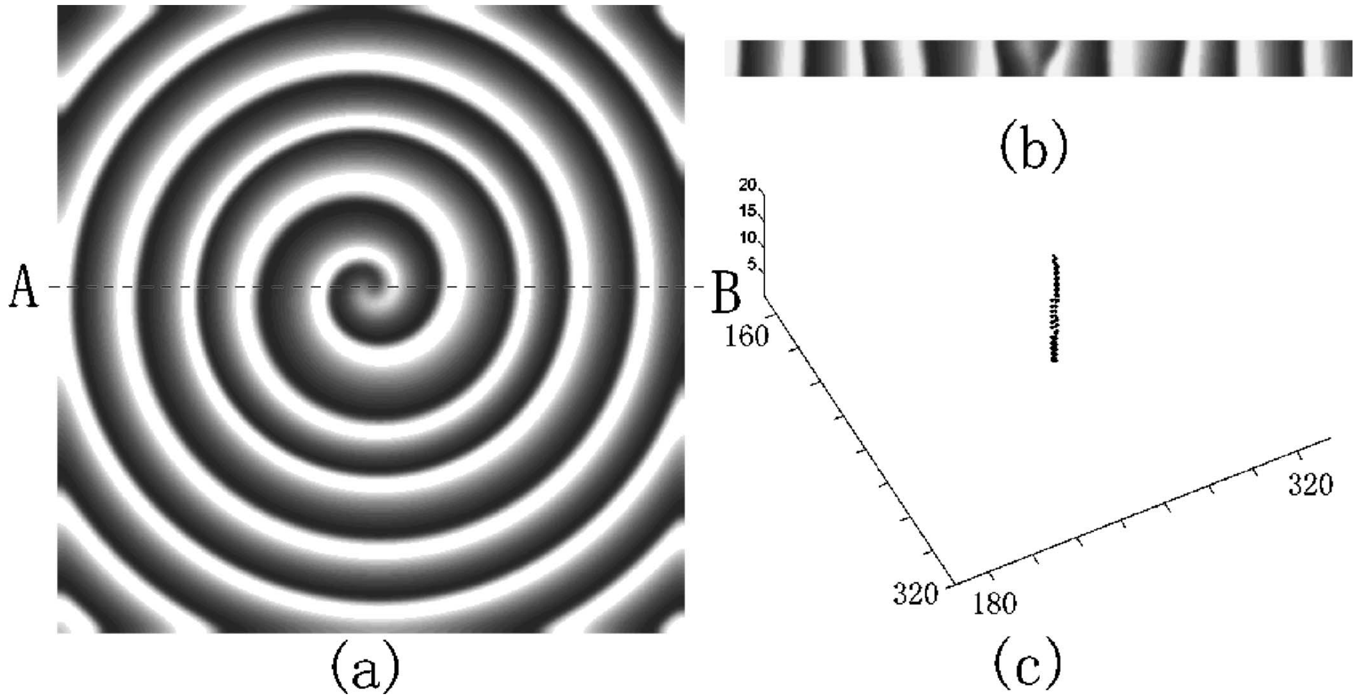

FIG. 1. Pictures of simple scroll waves with parameters set at $a_1=-0.154$. (a), 2D projection of scroll wave; (b), vertical cutting plane of line A-B in (a); (c) filament picture of the whole space: all defect points are signed by black points. In (a) and (b), dark areas stand for low values of u; light stands for high values of u.

and v. The dynamic equations are the following:

$$\frac{\partial u}{\partial t} = f(u,v) + \nabla^2 u, \quad \frac{\partial v}{\partial t} = g(u,v). \tag{1}$$

The local reaction kinetics are given by

$$f(u,v) = u(1-u)(u-a) - v, \quad g(u,v) = \varepsilon(u-v), \tag{2}$$

where $\nabla^2 = \partial^2/\partial x^2 + \partial^2/\partial y^2 + h\partial^2/\partial z^2$, the diffusion of v is omitted; ϵ and a are parameters. The parameter ϵ is small so that the time scale of u is faster than that of v. The parameter a is chosen as the control parameter in the simulation. The calculations were done in the Cartesian coordinate system with grid size $512\times512\times20$ segments, which is equivalent to coupling 20 different layers of the 2D system together and mimics the reaction medium in our experiment. The intensity of the coupling can be adjusted by choosing different h. The simple Euler method is applied with a time step $dt=0.03$ time unit and a space step $dx=dy=dz=1$ length unit.

In the simulation, we let a be homogeneous in the X and Y direction and vary at different Z. A series of $a_1, a_2, a_3, \ldots, a_{20}$ is used to describe the local value of control parameter a in each layer. a_1 represents the bottom layer and a_{20} represents the top one. Since it is easier to increase mean concentration than to increase gradient in experiments, we set the series a an arithmetical progression and keep $a_{i+1} = a_i + 0.001$. We change all a values uniformly so that a_1 is enough to represent all a values of the whole system. The system was studied under no-flux boundary conditions, and other parameters are fixed at $\varepsilon=0.08$, $h=0.32$, unless otherwise indicated.

The control parameter is selected such that the whole system is in an oscillatory regime. We concentrate on studying the asymptotic behavior of the system. For each set of control parameters, we let the system evolve for enough time (7500 time unit) to make sure that it attains to its asymptotic state. We should emphasize that within the whole parameter region of our simulation, spirals in the 2D system are always stable so that the spiral breakup in the simulation is due to a 3D effect, more specifically, the gradient effect.

III. RESULT

In a 2D system, a spiral pattern becomes unstable via a long wavelength instability as the control parameter a increases beyond -0.127. In the following, we keep a under -0.127 so that the system is always stable in a 2D system. In the 3D medium, the scroll wave is stable when the parameter $a_1 \leqslant -0.154$. If we observe it from the Z direction of the reaction medium, as observers do in most experiments of the reaction-diffusion system, we get a 2D projection by adding concentrations along the Z direction. The result is shown in Fig. 1(a). It suggests that the whole vortex can be legitimately viewed as a stack of 2D spirals, because the waves in the third dimension are well entrained [22] and instabilities in this dimension could be avoided. In this case the coupling strength (the effect of diffusion among layers) is strong enough to drive the whole vortex running together. In order to show the configuration of the scroll wave from another visual angle, we collect data from a vertical plane which cuts through the central filament (line of defects). When the system is rotational symmetric, information of such a cutting

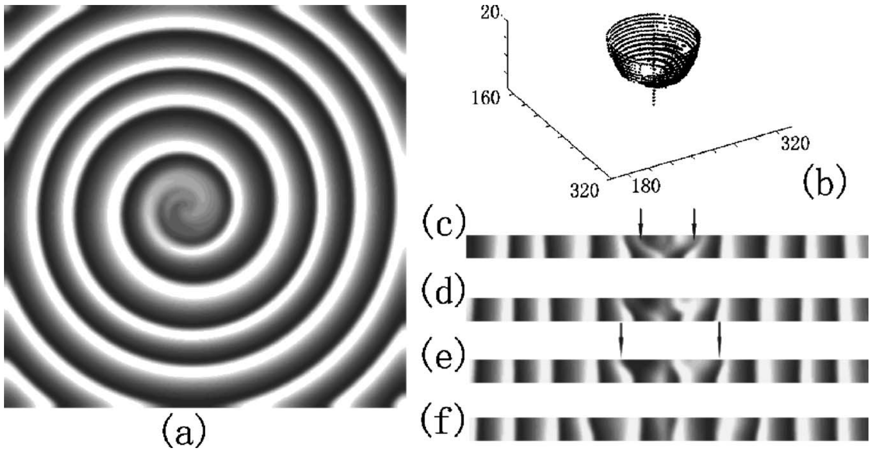

FIG. 2. Phase-shift phenomenon of the phase-twisted scroll wave. (a) 2D projection at $t=14\,292$; (b) filament picture at $t=14\,292$; (c) wave fronts are crowded at the arrow point, $t=14\,292$; (d) a wave front is shattered and a phase twist is eliminated by 2π at $t=14\,373$; (e) Last front (pointed by the arrow) is excessively inclined at $t=14\,457$; (f) the scroll recovers to a simple untwisted status at $t=14\,880$.

plane is enough to describe the whole system. As expected, we see 2D waves propagating outward on the cutting plane; a snapshot of such waves is shown in Fig. 1(b). Due to the gradient of the control parameter a, different layers of the system will have different oscillation frequencies if they are decoupled. This difference in intrinsic frequency makes the wave fronts near the center become inclined instead of vertical, which means that the vortex has phase differences along the vertical direction; it is named "phase twist" [18]. We will see in the following that this phase twist makes the system less stable, causing scroll ring formation and finally leading to a transition to spatiotemporal chaos. In addition, we single out all the defects in the space and get a filament picture of the system, see Fig. 1(c), in which only one straight vertical filament could be seen. Here, our definition of defects is $|u-u_0|<u_\varepsilon$, $|v-v_0|<v_\varepsilon$, in which $(u_0,v_0)=(0,0)$ is a fixed point calculated from local dynamics and u_ε, v_ε are sufficiently small values.

Although the scroll wave seems like a stack of 2D spirals for most of time when $a_1\leqslant-0.154$ as in Fig. 1(a), it shows periodic instable signals. Every once in awhile in the central area (about a wavelength away from the filament of the scroll wave), the wave becomes vague for awhile before it returns to be clear again. A snapshot of the vague pattern is shown in Fig. 2(a). This phenomenon occurs repeatedly with a period of tens of scroll wave cycles. In 3D defects picture, one observes temporary new born defects in the same region, which compose a shape like a cyclone, as shown in Fig. 2(b). Since the spontaneous creation of defects is a signal of a wave breakup, the system is undergoing periodic breakups and recoveries. During these breakup and recovery cycles, we observe on the cutting plane that the phase twist of the vortex is developing all the time. With the persistent accumulation of phase differences, the wave front becomes more and more inclined. When the wavelength of upper layers becomes too small for the system to support, the wave front breaks up in these layers. Figure 2(c) gives a snapshot at the moment of the wave breakup.

Unlike in 2D systems, such a breakup does not definitely lead to the destruction of the whole vortex. As we observed in the simulation, the cyclone-shaped defects disappear as a function of time; the system returns to its stable status. We notice that when the breakup happens along a wave front, the phase difference between the two sides of the breaking front is eliminated by 2π, so that the physical effect of this partly breakup and recovery is actually a phase shifts that releases the phase twist accumulated with the elapse of time [see Fig. 2(d)]. Although phase shifts enable the following wave fronts to become vertical, the last tilted wave front that produced before the phase shift (which we will call last front in the following text) is still propagating outside. Usually tilted wave fronts gradually become vertical as traveling outwards. In the meantime, we notice an anomaly that last front tends to become particularly tilted and shows a potential of breakup during a short period of time after the phase shift finishes, as shown in Fig. 2(e). However, when the control parameter is below $a_1=-0.154$, the last front ultimately becomes vertical at the end of such a break and recovery cycle, as shown in Fig. 2(f). Generally speaking, except for periodic phase shifts, the scroll vortex is stable.

As the parameter a_1 goes beyond -0.154, the last front becomes so tilted that it introduces a permanent scroll ring. In this case, the wave breaks up at about two wavelengths away from the center, which is resulted from too small a local wavelength on the top layer, as shown in Fig. 3(a). When this happens, defects appear in the top layer at the beginning, forming a line of a closed scroll ring. Unlike the temporal cyclone-shaped defects shown in Fig. 2(b), this scroll ring does not disappear after its formation. Instead, the ring expands a little to a balanced diameter, moves downward, and settles in about the middle layers of the medium, see Figs. 3(b) and 3(c). Because the phase of the waves emitted from the central filament varies with an angular position (at a fixed radius), the scroll ring is intrinsically twisted. One can get such a once-twisted scroll ring by twisting a straight scroll 360° and bending it, then sealing its ends together to preserve continuity (see Fig. 3c in [13] for an example). The waves that the scroll ring generates collide and keep balance with the waves generated by the central straight filament, while rotational symmetry is persistently kept. A series of cutting plane pictures forming a rotation cycle are given in Fig. 3(d), which show the movement of the scroll ring and its waves. In our simulation, this complex but symmetric type of scroll wave pattern is asymptotically stable in a considerable parameter region. Given that the system still undergoes periodic phase shifts, and since the established scroll ring expands away from central area where the ring is born, periodically, a new small scroll ring emerges in much the same

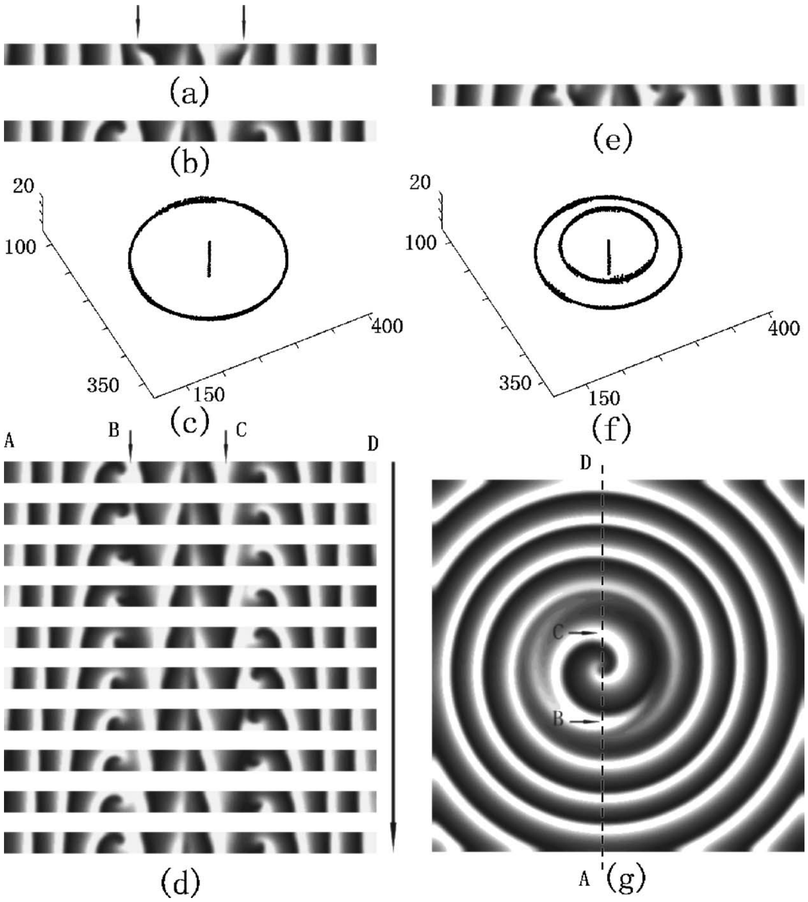

FIG. 3. Creation of scroll ring. (a) Last front becomes so inclined that a breakup happens at $t=15\,648$; (b) the projection of the scroll ring in the cutting plane shows a couple of spirals at $t=16\,761$; (c) defects of the scroll ring in 3D space at $t=16\,761$; (d) the evolution of the complex scroll pattern in a rotation cycle of the scroll wave ($t=16\,761-16\,791$). (e) and (f) the new born small scroll ring ($t=17\,805$); (g) 2D projection of the scroll ring pattern of (b), (c), and (d)1, in which the cutting plane is shown by the dashed lines. The arrows B and C in (d)1 and (g) indicate the corresponding points in the two pictures with conspicuous whiteness. The arrows point at areas inside and close to the scroll ring in (d)1 so that they help locate the position of scroll ring in (g) (which should be a little outside the B and C areas).

way as the former one, as shown in Figs. 3(e) and 3(f). The new ring moves down at first but then returns towards the top boundary and disappears. When a scroll ring exists, the spiral pattern in a 2D projection shows quite distinctive characteristics at the corresponding region of the ring: a thick and luniform wave arm, a fissure along the wave arm as well as a light and shaded contrast near it appears. Figure 3(g) presents an example of such a pattern. More than one ring corresponds to more than one luniform wave arm.

When we continue to increase a_1 to -0.152, the new born small ring in Fig. 3(f) tends to incline away from the X-Y plane so that part of it remains in the system while the other part may go out of the top boundary. Such unclosed filaments have two ends at the top boundary and are called "hot-dogs" [6]. An example of such a structure in a 3D space is shown in Fig. 4(a). Although hot-dogs can drift upward and disappear, rotational asymmetric effects are introduced into the system. More specifically, the phase gradient along the Z direction will vary with angular positions. After this symmetry breaking event, when the phase twist accumulates to the threshold for a phase shift to occur, due to the disaggregation of the rotational symmetry of the phase gradient, the wave breakup can never occur at all directions simultaneously. The defects that used to develop into a whole cyclone now appear only in a certain direction. They move toward the central filament and interact with it [Fig. 4(b)], drag it to bend, and greatly affect the phase twist of other direction. After that, hot-dogs continue appearing irregularly, which indicates the state of turbulence; see Figs. 4(c) and 4(d). The corresponding 2D projections are presented in Figs. 4(e) and 4(f), which show characteristics of a spiral breakup at one wavelength away from the center.

Such spiral breakup phenomena were observed in our experiment, which was conducted in the same spatial open reactor described in earlier studies [4,5,17]. The reaction medium is a porous glass disk, 0.4 mm in thickness and 25 mm in diameter, which is in fact sub-three-dimensional. Contacting each surface of the membrane with a reservoir, where reactants are refreshed continuously, fulfills a homogeneous and constant boundary condition of the reaction medium. The ferroin catalyzed BZ reaction is used in the experiments. The chemicals are arranged in two reservoirs in such a way that one (I) is kept in the reduced state of the BZ reaction, the other (II) is kept in the oxidized state. So that there are chemical gradients across the Z direction of the reaction medium. In our previous studies [3–5], boundary conditions were carefully chosen such that the patterned layer is thin, so that the waves in the third dimension are well entrained [22] and instabilities in this dimension could be avoided. Here, we deliberately increase the thickness of the patterned layer so that dynamic instability in the gradient direction must be considered.

From the snapshot picture of Fig. 5(a) taken from the experiment, we could guess the existence of complex 3D structures. Although it is hard to illustrate it in the experiment, the characteristics of the patterns still give us certain indirect evidence. One observes in Fig. 5 that the white areas in both the experiment and simulation have similar luniform

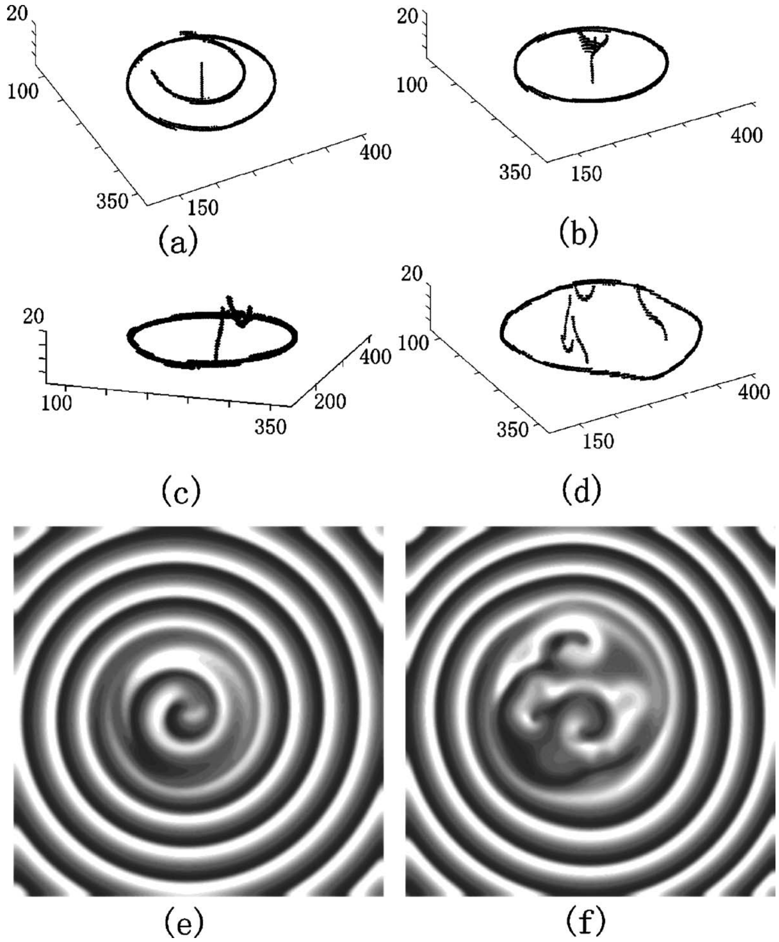

FIG. 4. Destabilization of the scroll wave. (a) small rings become inclined and broken at $t=25\,779$; (b) asymmetric cyclone contacts the central filament at $t=26\,262$; (c) irregular hot-dogs emergence at $t=26\,472$; (d) defects mediated turbulence at $t=29\,382$. (e) and (f) are 2D projections of (c) and (d), respectively.

shapes. From the comparison between Figs. 3(d) and 3(g), we believe that there may be scroll rings at points 1–3 in Fig. 5(a) and points 4 and 5 in Fig. 5(b). This experimental result qualitatively supported the existence of scroll rings in a medium with the gradient. However, a discrepancy exists. Unlike typical patterns in simulations, which primarily show one scroll ring [like in Fig. 3(g)], patterns in the experiments always contain several scroll rings spreading from near the center to many wavelengths away. That is probably because there are inhomogeneity in the X-Y plane in the experiments, which welcomes more scrol ring formation; or such a gradient effect is due to a long temporary process before the system attains to the asymptotic state. In the simulation, if we extend our attention to all the temporary process, patterns with pronounced multirings resembling that observed in the experiment [Fig. 5(a)] could be found. Figure 5(b) gives such an example in the simulation. Furthermore, before the scroll ring's creation, periodic change in the tip area is also observed in experiments which mimics phase shifts in simulation. Figure 5(c) and 5(d) ($a_1=-0.154$) show similar thick tips (near points 6 and 7, respectively) of a 2D projection at an earlier stage of scroll ring formation, which indicate the inclination of the wave front as a result of the 3D effects.

IV. DISCUSSION AND CONCLUSION

As mentioned at the beginning of Sec. III, within the whole parameter region of our simulation, spirals in the corresponding 2D system are stable. Although the region is near the B-F-N boundary, the phenomenon of destabilization at about one wavelength away from the center is completely different from long wavelength instability in which defects permeate from the periphery of the spiral. Also, filaments never show negative tension during the whole process. Even after the destabilization, straight filaments can spontaneously recover after it is "dragged" by other filaments.

Although the space step we used ($dx=dy=dz=1$, effective $dz\approx 1.7$) is larger than the conventional value $dx=0.3-0.5$ in simulations, we have confirmed that the result is not due to numerical discretization. We have repeated the scroll ring's creation using $dx=dy=0.5$, $dz=1.0$, and $dx=dy=1.0$, $dz=0.5$, respectively, both reaching exactly the same phenomenon. Taking account of the wavelength (larger than 30) and the slope of the wave front (not as sharp as in excitable medium), our space step is acceptable. Extra simulations in a 2D homogeneous medium with a typical parameter $a=-0.135$ were performed to prove the robustness of numerics: we get spiral waves with approximately the same wavelength and frequency with dx ranged from 0.4 to 2.8. When dx goes above 2.8, the simulation fails and presents chaotic patterns. So the space step we used above is justified.

We believe the spontaneous creation of scroll rings and hot-dogs are the cause of the final spiral instability. The direct cause of wave breakups and scroll ring or hot-dog formation is that the local wavelength reaches the minimum limit determined by the dispersion relation [23]. In our simu-

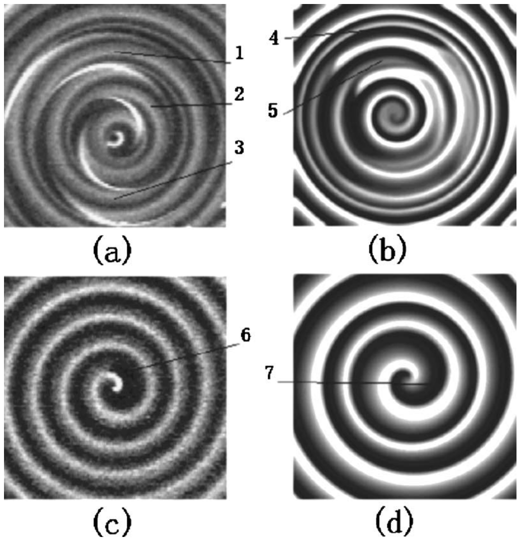

FIG. 5. Comparison of experimental observations and simulations. (a) spirals with possible scroll rings observed in experiments; (b) projection pattern of scroll waves soon after a_1 changes from -0.155 to $-0.154(h=0.50)$. The control parameters in the experiment are (a) $[H_2SO_4]^I=0.8$ M, (c) $[H_2SO_4]^I=0.6$ M. The other parameters are kept fixed: $[NaBrO_3]^{I,II}=0.3$ M, $[KBr]^I=60$ mM, $[H_2SO_4]^{II}=0.2$ M, $[Ferroin]^{II}=1.0$ mM.

lation, the phase shift (Fig. 2) is such a breakup that occurs periodically. Because the gradient of the control parameter along the Z direction brings a frequency difference, phase twists around the central filament accumulate as a function of time. When the phase twist is so large that wavelengths of upper layers reach the minimum limit, a breakup happens.

The most interesting phenomenon here is the spontaneous creation of a scroll ring and its coexistence with the central straight filament [see Fig. 3(c)]. Some topological analysis of this new-born filament is helpful in understanding the scroll ring structure. Generally any breakups occurring inside the medium should create a pair of defect points (2D situation) or a pair of filaments (3D situation) to conserve the topological charge. However, if the breakup occurs at the boundary (under no-flux boundary condition), the situation is different. In this case, tips (2D situation) or filaments (3D situation) coming from outside the boundary could be single instead of in pairs. To explain why this scroll ring occurs symmetrically, we argue that the breakup observed in this simulation originates from the gradient of oscillation frequency. It is a regular and rotational symmetric force responsible to the phase-twist accumulation and wavelength reduction at every angular direction. In every cutting plane, local wavelength at a definite point [such as pointed by arrows in Fig. 2(c)] will be persistently reduced until a breakup occurs. So when the phase twist reaches a critical point, deterministic breakup occurs, which is identical to every angular direction. Although phase gradients at different angular directions may be slightly different due to fluctuations, which may cause waves to break at some definite direction a little earlier or later, we believe that such an effect can be neglected in a large time scale (several spiral cycles) so that it does not contradict with the creation of symmetric filaments.

The appearance of cyclone-shaped defects during the phase shifts are incomprehensible in traditional asymptotic scroll wave patterns, because it seems like a 2D structure instead of a few 1D filaments. However, we think that temporary defects cannot be necessarily viewed as organizing centers of scroll waves. The cyclone-shaped defects, which only exist for no more than one rotation cycle of the scroll wave, just represent the appearance of a curved surface with zero temporary oscillation amplitude. During a phase shift, the oscillation amplitudes of all the points at that surface decrease to zero so that their relative phases could be instantaneously adjusted. After that, the oscillation amplitudes return to normal values and the cyclone quickly disappears.

In order to verify the relation between the frequency difference along the central filament and the creation of new filament, we did some simulations in 2D homogeneous medium to learn the characteristics of 2D spirals under different parameter a. The result is summarized in Fig. 6(a). Since the

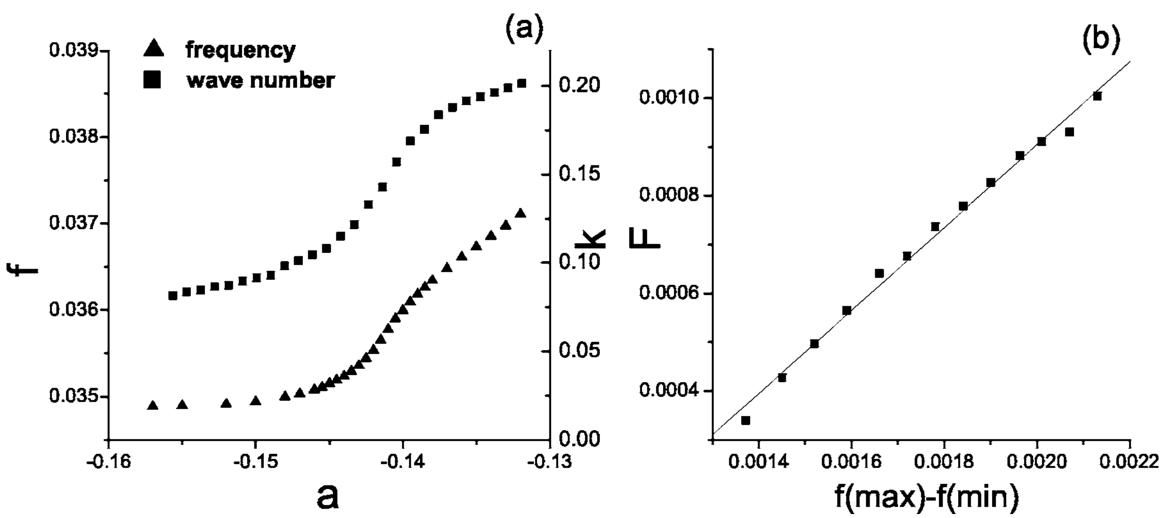

FIG. 6. (a) The rotation frequency f (triangle) and wave number k (cube) of the 2D spiral wave system measured at $r\rightarrow\infty$, as a function of the control parameter a. (b) The frequency of phase-shift F measured as a function of the frequency difference between top and bottom layers calculated from (a).

relation between the control parameter a and intrinsic rotation frequency f is nonlinear, when the control parameter a_1 in our 3D simulation varies, the oscillatory frequency differences among layers also change. Correspondingly, some properties of filament creation will change. This may explain the different behavior of the system when a_1 varies. Figure 6(b) illustrates an approximately linear relation between the frequency of phase shifts in 3D simulations and intrinsic oscillatory frequency differences between the top layer and the bottom layer. Obviously, coupling strength among layers minimizes the frequency difference. When the intrinsic frequency difference is small enough, the phase shift does not happen and the phase twist is fixed. This is just the stable case reported in prior documents, [8,9,16] and is also observed in our simulation when $a_1 < -0.160$.

Finally, as the control parameter a_1 continuously increases, the frequency difference between the top and bottom layers of the system attains a critical point; the scroll ring that was created from the wave front breakup becomes asymmetric, giving hot-dog-shaped filaments, inducing the system to undergo a transition to defect-mediated turbulence. The underlying mechanism of this rotational symmetry breaking is still a question for further investigation. Here we give some elementary proposal for explanation. Since the form of this scroll pattern solution is too complicated for linear stability analysis, we resort to a method of filament dynamics [6,7]. It was stated that a scroll ring may turn its normal to the direction of the parameter gradient, which is the reason why the big scroll ring could exist horizontally in the system. We suppose that the vortex with the phase twist originated from the central filament imposes forces on the scroll ring, whose amplitude and direction vary with the distance from the center and the height (Z position). The interaction may give the scroll ring a torque and drive it inclined.

In conclusion, we observe a spontaneous scroll ring and hot-dog formation followed by a new type of scroll instability in our simulation, and carefully examine each stage of its process towards destabilization. We find that the gradient of the rotation frequency is an original cause of the spontaneous creation of filaments, while the spontaneous creation of asymmetric filament is the direct cause of the scroll's destabilization. Since our finding is confined in the oscillatory regime, whether such kinds of instability also exist in the excitable medium remains a question. Finally, we should point out that our analysis about symmetry breaking in this system is elementary. To develop a systematic theory of this type of instability, a mathematical analysis of complex patterns is needed.

ACKNOWLEDGMENTS

We thank Y. Li and C. Fang for their elementary simulation result; A. S. Mikhailov, F. Sagues, and S. Alonso for sharing their ideas and helpful suggestions. This work is partially supported by the Chinese Natural Science Foundation and the Department of Science and Technology in China. C.W. and S.W. thank the Jun-Zheng Foundation at Peking University for support. C.W. and S.W. contributed equally to this work.

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