Excitable media support the transmission of propagating waves, which typically spread radially from a point source or organize themselves into rotating two-dimensional spirals or three-dimensional scrolls. Spiral waves have been extensively studied in the Belousov–Zhabotinsky (BZ) reaction and in living systems such as the slime mold Dictyostelium discoideum. Scroll waves have also been extensively studied in the BZ reaction as well as in migrating Dictyostelium slugs. Many aspects of spiral and scroll wave dynamics can be accounted for in terms of eikonal equation descriptions. Studies of scroll rings by Pertsov and co-workers have shown that the evolution of such rings is strongly influenced by an excitability gradient arising from a temperature gradient across the medium. They have also studied the destabilization of linear scroll filaments in temperature induced excitability gradients. We study in this paper the formation and evolution of scroll waves in the photosensitive ruthenium-catalyzed BZ reaction. The photosensitive BZ system allows the generation of precisely oriented scroll rings, permitting the study of their evolution in light-induced excitability gradients of varying intensity and direction.

I. INTRODUCTION

The excitability of the photosensitive BZ system can be readily controlled by illumination with visible light. Bromide, which is an inhibitor of autocatalysis in the BZ reaction, is produced in a photochemical cycle with exposure to 460 nm light. This, in turn, diminishes the activity of the medium from oscillatory to excitable to nonexcitable, depending on the light intensity and composition of the medium. The excitability transverse to the direction of wave propagation can be controlled by irradiating a thin gel medium from below. A scroll wave can be initiated from a two-dimensional traveling wave by perturbing the excitability in the transverse direction, thereby changing the character of the medium from effectively two dimensional to three dimensional. The initiation technique is similar to that first used by Winfree and co-workers, and later by Krinsky and co-workers and Linde and Engel, in which the thickness of a wave-filled medium is suddenly increased in the transverse direction by adding new excitable solution or gel.

Experimental and modeling results on scroll wave initiation are described in Sec. II and the evolution of these waves in the presence of excitability gradients is described in Sec. III. We discuss our results in the context of previous studies in Sec. IV. Experimental methods and procedures are given in the Appendix.
FIG. 1. Formation and evolution of perturbation-induced scroll wave. Successive subtraction images (with 1.0 s between primary images) show wave behavior at 5.0 s intervals in a silica gel medium 0.5 mm thick. White and black correspond to wave front and wave back, respectively. The medium was exposed to low-level homogeneous illumination from below using an integrating sphere and quartz-halogen light source (irradiance: 6.56 mW cm\(^{-2}\)). The first frame shows a developing target pattern 10.0 s after its initiation with a silver wire. The system was perturbed with high-intensity illumination (irradiance: 9.64 mW cm\(^{-2}\) from \(t=16\) to 24 s. Image area: 15.2 mm \(\times\) 17.0 mm. Composition of the BZ solution is described in the Appendix.

period of 8.0 s. Although the target pattern seems to expand unperturbed, as shown in panels (d) and (e), a wave induced by the perturbation appears in the wave back of the parent wave after a short delay, as can be seen in panel (f). The new wave proceeds to split into two distinct wave fronts, one of which propagates inward while the other propagates outward, panel (g). This behavior is repeated with the periodic appearance of new circular wave sources, panels (h), (k), and (n), which also split into inward and outward propagating circular waves.

The wave sources arise from a scroll wave within the thin rectangular gel, which strikes the surface of the medium as it rotates around its circular filament (vide infra). The space–time plot in Fig. 2 shows the initial appearance of the wave source and its successive reappearances (at the local minima in each of the curves). This plot was constructed from gray levels along the horizontal section shown by the dashed line in panel (a) of Fig. 1, which passes through the initiation point of the original wave. The waves propagating toward the center of the medium are the contracting circular waves which self-annihilate at the initiation point. The waves propagating outward are the expanding circular waves. We note that the periodic wave source expands in time, indicating an expanding scroll ring filament. Over 20 rotational periods of the scroll wave were observed in this particular experiment.

B. Modeling

The Tyson–Fife scaling of the Oregonator\(^{25,26}\) modified to describe the photosensitive BZ reaction\(^{22,24,27}\) was used to simulate the scroll-wave behavior:

\[
\frac{\partial u}{\partial \tau} = \delta \nabla^2 u + \left(1/\epsilon^3\right) \left( q w - u w + u - u^2 \right),
\]

\[
\frac{\partial v}{\partial \tau} = u - v,
\]

where \(u, v,\) and \(w\) are the dimensionless concentrations of HBrO\(_2\), Ru(bpy)\(_3^+\), and Br\(^-\), respectively. \(\nabla^2 = (\partial^2/\partial x^2 + \partial^2/\partial y^2 + \partial^2/\partial z^2)\) is the Laplacian operator, and \(\delta = D_u/D_w\) is the ratio of the diffusion coefficients. We let \(\delta = 1.0\) and set \(D_w = 0\) for the immobilized Ru(bpy)\(_3^+\) catalyst. The rate of bromide production from the irradiation is given by \(\varphi\), \(\epsilon\), \(\epsilon^3\), and \(q\) are scaling parameters, and \(f\) is an adjustable stoichiometry parameter.

We consider illumination of the medium in the \(z\) direction from above or below. The perturbation by high-intensity light for initiating scroll wave activity is always applied from below. Because the light is absorbed as it passes through the medium, the illumination introduces a Br\(^-\) concentration gradient in the \(z\) direction. The light induced Br\(^-\) production is attenuated according to a Beer–Lambert relation,

\[
\varphi(z) = \varphi_0 \exp(-\alpha z),
\]

where \(\alpha\) includes the molar absorption coefficient and concentration of the reduced catalyst, Ru(bpy)\(_3^+\). \(\varphi_0\) is the quantum efficiency for the photochemical production of Br\(^-\), and \(z\) is the dimensionless distance from the surface of the medium exposed to the incident light. The calculation was
simplified by treating $\alpha$ as a constant parameter, with the photoinduced bromide generation dependent only upon the value of $z$.

The finite difference form of Eqs. (1)–(3) was integrated in a three-dimensional grid ($N_x \times N_y \times N_z = 321 \times 321 \times 48$) by an explicit Euler method ($\Delta t = 1/3$, $\Delta z = 1.3888 89 \times 10^{-3}$) with no-flux boundary conditions along the perimeters. The time step $\Delta t$ was judged to be sufficiently small by determining when the behavior remained the same with smaller time steps. The Laplacian terms in Eqs. (1) and (3) were approximated by using the six nearest grid points according to the standard seven-point formula for three-dimensional media.

The mechanism for the scroll wave formation can be seen from the numerical integration of Eqs. (1)–(3) in Fig. 3. The $x,z$ images show the behavior along a horizontal cross section (indicated by the dashed line in the first panel) of the corresponding $x,y$ images. Panel (a) shows the newly formed wave, which was initiated at the top of the medium and has yet to fully reach the bottom. Increasing the light intensity during the perturbation period in panel (b) causes the formation of free wave ends due to the decrease in excitability in the $-z$ direction. When the original, lower light intensity is restored in panel (c), the free ends curl to form the filament of a circular scroll wave, as shown in panels (d)–(f). The wave source arises from the scroll wave striking the upper surface of the medium, as shown in panel (g), thereby generating inward and outward propagating circular waves, panel (h). The wave front of the inward propagating wave is more pronounced than that of the outward propagating wave, much like the experimental behavior shown in Fig. 1.

III. EVOLUTION OF SCROLL WAVES

A. Contracting scroll filaments

Scroll waves generated by perturbing circular waves propagating inward rotate in the opposite direction relative to those generated by perturbing circular waves propagating outward. The technique for producing such waves involves an extension of the method described above. As shown in Fig. 4, a target pattern is perturbed to form a scroll filament, panel (a), with the subsequent appearance of inward and outward propagating daughter waves, panel (b). Now, however, the system is perturbed again, panels (c) and (d), to form scroll waves from each of the daughter waves as well as the original parent wave. After restoring the low-intensity illumination, images show behavior at $t = 3.1$ (a), 4.9 (b), 6.6 (c), 8.3 (d), 10.1 (e), 11.8 (f), 13.5 (g), and 15.3 (h).

FIG. 3. Three-dimensional simulations of the perturbation-induced scroll waves using the modified Oregonator model, Eqs. (1)–(4). Images were obtained by subtracting two primary images giving the concentration of $u$ ($\Delta t = 0.35$ between primary images), with white assigned to the highest value in the wave front and black to the wave back, respectively. Two images are shown for each sampling time: the upper $x,y$ image shows the top view as in Fig. 1; the lower $x,z$ image is the middle cross section indicated by the dashed line in the first $x,y$ image. The aspect ratio of the images is $x:y:z = 1:1:2$. The perturbation of high-intensity illumination was applied from below from $\tau = 4.2$ to 5.2. Parameters of Eqs. (1)–(3): $\epsilon = 6.61 \times 10^{-2}$, $\alpha = 3.67 \times 10^{-2}$, $q = 9.52 \times 10^{-4}$, $f = 1.05$ (Ref. 24). Parameters of Eq. (4): $\alpha = 0.232$, $\varphi_0 = 7.20 \times 10^{-3}$ for low-intensity illumination and 0.180 for high-intensity perturbation. Images show behavior at $t = 3.1$ (a), 4.9 (b), 6.6 (c), 8.3 (d), 10.1 (e), 11.8 (f), 13.5 (g), and 15.3 (h).

FIG. 4. Wave evolution arising from two subsequent perturbations. Image areas: 15.3 mm$\times$13.4 mm. Snapshot interval is 4.0 s and perturbation periods are from $t = 20$ to 26 s and from $t = 44$ to 51 s. Other conditions and procedures are the same as in Fig. 1.

The space–time plot shown in Fig. 5 illustrates the distinct phases of this experiment. The formation of the initial scroll wave following the first perturbation can be clearly seen, as well as the subsequent formation of three scroll waves, P1, P2, and P3, after the second perturbation. Of the two surviving scroll rings, the one arising from the outward propagating daughter wave expands (P2), while the one arising from the inward propagating daughter wave contracts (P3). Figure 6 shows a space–time plot of a similar contracting scroll wave in a longer duration experiment in which five scroll rotations can be seen.
B. Dependence on excitability gradient

Scroll wave filaments are known to shrink and eventually collapse in isotropic BZ media. Our experiments show that in an excitability gradient produced by light, scroll filaments may shrink, as in Fig. 6, or expand, as in Fig. 2, depending on the gradient strength and the direction of the scroll wave rotation relative to the gradient. Experiments were carried out to examine the effect of the illumination intensity on the evolution of the scroll filament. Circular waves propagating inward or outward were perturbed by high-intensity illumination to generate scroll waves with different rotational directions. The results of these experiments are summarized in Fig. 7, where the scroll filaments arising from expanding and contracting circular waves are indicated, respectively, as diamonds and squares. A scroll filament formed by perturbing an expanding circular wave contracts in the presence of a small excitability gradient at very low-intensity illumination. This is in agreement with earlier studies showing that scroll filaments contract in gradient-free BZ media. On increasing the illumination intensity, however, the contraction rate decreases, and, at a critical intensity, the filament is stationary. Further increasing the light intensity causes the filament to expand: the higher the intensity, the greater the expansion rate. An upper limit occurs at a light intensity where the medium no longer supports scroll wave rotation. In contrast to this behavior, scroll wave filaments arising from the perturbation of an inward propagating circular wave always contract, with the contraction rate increasing with increasing light intensity, as shown by the squares in Fig. 7.

Numerical simulations were also carried out to determine the effects of illumination intensity and relative direction on the scroll wave evolution. The space–time plots shown in Fig. 8 are analogous to the experimental plots in Figs. 2, 5, and 6, showing wave position at the surface of the medium as a function of time. Wave positions in the horizontal cross section [cf. panel (a) of Fig. 3] are shown, and the location of the scroll ring filament is indicated by the local minima of each curve. Panels (a) and (b) show scroll waves generated by perturbing outwardly propagating parent waves as in the experiments. For most illumination intensities, a steady increase in the radius of the scroll filament ring is exhibited, as shown in panel (a). The rate of ring expansion was found to increase with increasing $\phi_0$ until an upper limit was attained, where the thickness of the excitable region of the medium was not sufficient to support scroll wave rotation. Under dark conditions, where $\phi_0=0$ and there is no excitability gradient, the scroll filament shrinks and eventually collapses, as shown in panel (b). The radial velocity of the scroll filament changes sign at a value of $\phi_0$ between 0.0027 and 0.0080 for our parameter values. This “window” represents the range of values over which no measurable change in radius occurred during 15 rotations of the scroll: the scroll ring expands and contracts above and below this range. The change from expanding to contracting scroll filaments is in qualitative agreement with the experimental behavior observed at low light intensity (cf. Fig. 7).

The excitability gradient can be reversed by changing
the direction of the incident illumination. Calculations were carried out in which a perturbed outwardly propagating circular wave was subsequently illuminated from above at a low-level background intensity. Scroll filaments generated in this manner and illuminated from above always contract. An example of the behavior is shown in panel $c$ of Fig. 8. A comparison of panels $b$ and $c$ shows that the rate of contraction with illumination from above is significantly greater than that under dark conditions. As illustrated in Fig. 9, the evolution is dependent on the direction of the scroll wave rotation with respect to the direction of the illumination and, hence, the excitability gradient. The filament may expand or contract, depending on the illumination intensity, when the rotational direction is the same as the direction of illumination, as the $(+z)$-direction shown in (a) and (b). In contrast, the filament always contracts when these directions are opposite, as the $(-z)$-rotational direction and $(+z)$-illumination direction shown in (c). The opposing configuration was carried out in the experiments by perturbing an inwardly propagating parent wave and illuminating the scroll wave from below, as in Fig. 6. Because the medium size was restricted in the calculations, it was more convenient to perturb an outwardly propagating parent wave and illuminate the scroll wave from above. We note that these two methods give rise to equivalent configurations, where the illumination is in the opposite direction to the rotational direction of the scroll.

With the parameter values and system size of these calculations, the scroll ring filament exhibited very little drift in the $z$ direction. Calculations were also carried out to test whether the filament expansion and contraction were associated with any boundary condition influence. The behavior remained unchanged, provided that the filament was able to rotate freely, when the scroll filament was placed at different depths within the layer by changing the perturbation duration, the value of $\varphi_0$ during the perturbation, and the medium thickness $N_z$.

C. Broken filaments

The perturbation technique for generating scroll waves was applied to wave segments as well as symmetrical circular waves, as shown in Fig. 10. Panel (a) shows an arc of a circular wave formed by suppressing wave activity in a portion of a circular wave by irradiating that region with high-intensity light. Such a wave segment would normally form counter-rotating spirals at the two free ends. This behavior is inhibited, however, when the perturbation technique is applied to generate a scroll wave. Following the uniform perturbation, a crescent scroll wave appears in the wave back of the parent wave, as shown in panels (b) and (c), forming a pair of waves propagating in opposite directions away from the scroll filament. This can be seen in panel (d), where two of the three waves in the horizontal cross section represent those arising from the scroll filament and the third (at the right-hand side) represents the parent wave. The scroll wave

![FIG. 8. Space–time plots from numerical simulations with different illumination conditions. (a) Expansion of scroll ring with background illumination from below and $I_0=0.0072$. (b) Contracting scroll ring under dark conditions with $I_0=0.0072$. The data were collected along the dashed line in Fig. 3(a) from the third top layer ($N_z=3$) of the grid with sampling rate $1/(75 \Delta \tau)$.](image1)

![FIG. 9. Schematic representation of expansion and contraction of scroll ring filament depending on the direction of the scroll rotation relative to the direction of illumination. (a) Expansion of the filament ring for $I_0>I_{\text{crit}}$ when the directions are the same. (b) Contraction of the filament ring for $I_0<I_{\text{crit}}$ when the directions are the same. (c) Contraction of the filament ring for all $I_0$ when the directions of illumination and scroll rotation are opposite.](image2)
strikes the surface of the medium again in panel (e) to form another pair of waves in panel (g). The crescent scroll wave appears periodically in the location of the original parent wave, as in panels (b) and (e), and the free ends fail to form spirals because they represent the ends of the filament of the rotating scroll wave.

Calculations to examine the behavior of a scroll wave formed from a wave with a free end were carried out by perturbing a rotating spiral wave. These calculations were analogous to those described in Figs. 3 and 8 for simulating scroll ring behavior. As shown in Fig. 11, the spiral, perturbed in panel (b), becomes a quasistationary wave source as the scroll wave periodically strikes the surface, panels (g) and (m). The curved filament is not completely stationary, however, as it slowly decreases in length with each scroll wave rotation.

IV. CONCLUSION

The excitability of the photosensitive BZ medium can be readily perturbed in a direction transverse to a propagating wave by varying the illumination intensity. A perturbation applied to a circular wave, consisting of a uniform change in illumination intensity, creates a scroll wave which serves as a periodic wave source. The subsequent evolution of the scroll ring is influenced by the presence of a gradient in excitability and on its direction and strength. In our experiments, the excitability gradient is perpendicular to the scroll ring and its strength depends on the intensity of the illumination. The thin gel medium was illuminated from below and the relative direction of rotation of the scroll wave was determined by perturbing expanding or contracting circular parent waves. The scroll ring always contracts when the illumination and rotation directions are opposing, and the contraction rate increases with increasing gradient strength. When the incident illumination and scroll rotation are in the same direction, the scroll filament contracts at low intensity illumination and expands at higher intensity. Stationary scroll wave filaments occur only at a critical excitability gradient corresponding to the change in evolution of the ring from contraction to expansion. Although only contracting scroll rings have been found in experimental studies of isotropic BZ media, we note that theoretical studies indicate scroll rings may expand under some conditions in excitable media. In our simulations, the illumination gives rise to a gradient in the z direction according to a Beer–Lambert law for the production rate of bromide ion. Yoneyama et al. have carried out simulations of the photosensitive BZ medium by linearly varying the stoichiometric ‘‘f-factor’’ of the Oregonator model to introduce an excitability gradient.

We also find that semicircular wave segments with free ends fail to develop into counter-rotating spirals after an illumination perturbation. This type of behavior was previously observed by Linde and Engel in a two-phase liquid-gel experiment. In two dimensions, a rotating spiral wave is self-sustaining and can, in principle, persist indefinitely. When this wave is perturbed to become a scroll wave, however, the filament slowly contracts in the presence of a weak excitability gradient (Fig. 11). Presumably, such scroll segments could also expand with a sufficiently strong excitability gradient arising from higher intensity illumination.

In the thin gel layer of our experiments, the scroll wave filament is constrained to be coplanar with the medium and perpendicular to the excitability gradient. The excitability gradient generated from highly uniform illumination allows measurements of the response of the scroll waves to gradient strength and direction. Even though the medium is only 0.5 mm thick, it is sufficiently three dimensional to support long-lived scroll waves, which may undergo up to 20 rotations. This behavior required conditions corresponding to relatively rigid spiral rotation without significant meandering. The long-lived scroll behavior in very thin gels demonstrates that even in ostensibly two-dimensional excitable media, three-
dimensional scroll wave behavior is possible when the medium excitability is suitably perturbed.

ACKNOWLEDGMENTS

We thank the National Science Foundation (CHE-9531515), the Office of Naval Research, and the Petroleum Research Fund for supporting this research.

APPENDIX: EXPERIMENTAL METHODS

Silica gel media\textsuperscript{13,34} were prepared by gelling a solution of 10\% (w/w) Na$_2$SiO$_3$ and 1.0 mM ruthenium(II)-tris-2,2'-bipyridyl sulfate by acidifying with H$_2$SO$_4$. The gel was cast with a uniform thickness of 0.5 mm onto a microscope slide (thickness of 1.0 mm) by using a Plexiglas mold. The BZ solution, containing 0.1 M NaBr, 0.25 M NaBrO$_3$, 0.30 M malonic acid, and 0.49 M H$_2$SO$_4$ (prior to the bromination of malonic acid), was prepared with deoxygenated, doubly distilled water and stored under argon gas. The gel on the supporting microscope slide was placed into a petri dish, covered with 20.0 ml of catalyst-free BZ solution (to a depth of 2.5 mm), and allowed to equilibrate with the solution for 5 min. The covered petri dish was thermostated at 25.0 ±1.0 °C and purged with a slow flow of argon gas.

The gel was exposed to light from a 1.0 kW tungsten-halogen lamp passing through an integrating sphere (with an inhomogeneity of less than 0.1\% over the field) and a 350–500 nm bandpass filter. The irradiance was monitored with a Model S20MM power meter (ThorLabs Inc.). The transmitted light from the medium was monitored with a CCD video camera (Pulnix TM-7CN) through a 460 nm narrow bandpass filter. The video signal was recorded with an S-VHS video recorder and processed with an image-acquisition board (512×480 pixels, eight bits per pixel).

Spatially homogeneous perturbations of the excitability of the medium were carried out by increasing the irradiance to 9.64 mW cm$^{-2}$ for 8.0±1.0 s, during which the wave fronts became visibly thinner due to the inhibitory effect of the light.\textsuperscript{18–22} The perturbation intensity and duration were chosen such that wave activity was inhibited only in the bottom layer of the gel and wave behavior continued in the top layer. When the light intensity was returned to the reference level, wave activity resumed throughout the thin gel medium.

\textsuperscript{3}A. T. Winfree, Science 175, 634 (1972).
\textsuperscript{6}P. N. Devreotes, Science 245, 1054 (1989).
\textsuperscript{19}L. Kuhntert, Nature (London) 319, 393 (1986).
\textsuperscript{26}J. J. Tyson and P. C. Fife, J. Chem. Phys. 73, 2224 (1980).