

# Understanding the Patterns in the BZ Reagent

Paul C. Fife<sup>1</sup>

## 1. THE PHENOMENON

One of the more intriguing discoveries in chemical physics during the last 20 years was that of mobile patterns in the Belousov-Zhabotinsky reagent.<sup>(35,36,38,41-43)</sup> Foremost among these patterns are the expanding rings and rotating spirals often seen in thin layers of the mixture. It is felt that these structures, seen and studied so easily for this particular type of reactive medium, are in fact representative of quite wide-ranging phenomena in complex diffusive systems. Their prevalence has been surmised from the results of numerical experiments on various models, as well as from rigorous and formal mathematical results obtained for the simplest of the models.<sup>(1,11,12,14,15,19-24,26,37)</sup> The possibility that they reflect analogous phenomena in physiology has been suggested many times; there is experimental evidence of spirals in cardiac tissue, and there are theories to the effect that these spirals are connected with heart pathologies.<sup>(16,39)</sup>

All of this has stimulated the desire to understand the basic mechanism behind the observed dynamic patterns. (An even more fundamental exotic phenomenon is the tendency of the BZ reagent, even if well stirred, to oscillate in its chemical composition. It is clear, for example from the analysis presented below, that there is a close connection between oscillatory behavior and pattern-forming behavior, but the emphasis in this paper will be on the latter.) Since the BZ reagent is probably the simplest system supporting spatial patterns which can be studied in the laboratory, the first logical step was to understand them in that context. And, of course, the first part of that first step was to understand the chemical reactions taking place in the reagent. The chemistry is indeed complex, but by now the specific reactions which occur are fairly well known.<sup>(2-4,31)</sup> There is still much to be learned about the rates of these reactions, however.

---

<sup>1</sup> Mathematics Department, University of Arizona, Tucson, Arizona 85721.

In this paper I wish to outline briefly the steps which have led from basic knowledge about the chemistry to our present understanding of the mechanism behind ring and spiral patterns in the BZ reagent. These steps involve chemical modeling, mathematical modeling, and finally mathematical analysis. The process generally has been passage from complicated models to simpler ones by use of insight into the total chemical mechanism and by formal asymptotic analysis. My emphasis will be on the mathematical aspects of these processes. The end result has been models which retain the features mainly responsible for the appearance of the phenomena in question, and yet are simple enough that their analysis is straightforward and no longer obscure.

See Refs. 9 and 10 for further discussions of most of the material presented here.

## 2. OREGONATOR MODELS

A satisfactory picture of the chemistry has been obtained by careful experimentation, mainly by Noyes and various coworkers.<sup>(2)</sup> Once this knowledge had reached a high enough level, it was possible to characterize the essential features of the total reaction mechanism by a much simpler caricature or skeleton mechanism, which, it was proposed, would have the same qualitative dynamics as the real mechanism. The simplified proposed mechanism was named the Oregonator<sup>(4)</sup> and consists of a few reactions among three chemical species with variable concentrations. This reduction was a major step toward our understanding, because the mathematics of the Oregonator model is easier both to do and to comprehend.

Credence in the Oregonator skeleton was bolstered by its ability to reflect most of the exotic features observed in the lab, and moreover by the demonstration by Tyson<sup>(33)</sup> that it could be derived in a systematic way from larger more realistic systems. Nevertheless, there have recently been alternatives to the Oregonator proposed<sup>(29,30)</sup> on the basis of chemical considerations. As far as accounting for the phenomena of interest to us here, all these alternative models, together with the original Oregonator, do equally good jobs in reproducing spatial patterns. Therefore I shall not mention their differences further.

Mathematically, the dynamics of a well-stirred Oregonator reagent would be described by a set of three nonlinear ordinary differential equations. The spatiotemporal behavior of an unstirred layer of this reagent would be described by these same differential equations, with added diffusion terms in the form of spatial Laplacian operators acting on the concentration variables. Thus, one arrives at a system of three nonlinear reaction-diffusion equations.

### 3. PC SYSTEMS

In the Oregonator differential equations and reaction–diffusion equations described above, there appear a number of parameters representing various combinations of rate constants. These parameters are not completely known and may vary according to the specific recipe used in making the reagent. However, it is clear that one should expect them to have different orders of magnitude; and under certain realistic order of magnitude assumptions, it was shown by Tyson<sup>(32)</sup> that these equations can be further simplified by invoking a pseudo-steady-state hypothesis. What this hypothesis amounts to is a rescaling of the three unknown functions (the concentrations of the reactants), in terms of the parameters mentioned, in a way dictated to some extent by the nature of the equations. This rescaling exhibits the typical orders of magnitude of the three concentrations. One of them turns out to be much smaller than the other two; this leads to a steady-state approximation, which essentially says that the rate of production of this small species is also small, and can for purposes of the approximation be set equal to zero. Setting it equal to zero yields an algebraic equation which can be solved for this small rescaled variable in terms of the other two. In this way, the system of three equations is reduced to two.

This reduced system of two reaction–diffusion equations has, in a certain realistic parameter range, the form of what I have called a “propagator–controller” (PC) system. A (chemical) PC system is defined as a reacting and diffusing system which supports wave fronts in some chemical species (the propagator species), the velocity and wave forms of these fronts being modulated by the concentrations of other species (the controller species). In the reduced Oregonator system, there is only one propagator and one controller. The former is  $\text{HBrO}_2$  (its concentration will be denoted by  $u$  below) and the latter is a cerium ion in its oxidized state (its concentration will be denoted by  $v$ ).

The system has a sigmoidal nonlinearity and a small parameter  $\varepsilon \ll 0$ :

$$u_t = \varepsilon \Delta u + \frac{1}{\varepsilon} f(u, v) \quad (1)$$

$$v_t = \varepsilon \Delta v + g(u, v) \quad (2)$$

where the nullcline  $f(u, v) = 0$  has the following shape in the  $u$ – $v$  plane:

To see that this is in fact a propagator–controller system, observe that its lowest-order formal approximation is

$$f(u, v) = 0 \quad (3)$$

$$v_t = g(u, v) \quad (4)$$

As long as  $v$  lies in the interval  $\underline{v} < v < \bar{v}$ , (3) can be solved for  $u$  as a function of  $v$  in three ways, represented by the three branches in the nullcline shown in Fig. 1. The middle branch is unstable with respect to the kinetic equations and will be excluded from consideration on that basis; but the other two are feasible and will be represented by

$$u = h_{\pm}(v) \tag{5}$$

which replaces (3). Solutions of (4) and (5) with  $u$  discontinuous may exist, the points of discontinuity representing transition points where the relation between  $u$  and  $v$  switches between the  $+$  relation in (5) and the  $-$  relation.

It will be assumed that  $v$  is continuous. In fact, if, at time  $t=0$ ,  $u$  and  $v$  are both continuous in space, it can be seen easily from (4) that  $v$  will remain continuous, but that  $u$  may become discontinuous by being “forced off the upper or lower knee” of the nullcline in Fig. 1.

So at any one instant of time, space will be divided into a region  $\Omega_+(t)$ , in which  $+$  holds in (5), and  $\Omega_-(t)$ , where  $-$  holds. Suppose the boundary between these two regions is smooth with curvature not large, i.e., small compared to  $1/\epsilon$ . Then a fine-structure analysis of the original system (1), (2) near the boundary<sup>(6,7,9,27)</sup> actually shows that the boundary must migrate, its normal velocity  $c$  being an  $O(1)$  quantity depending only on the value of  $v$  at that point:

$$c = c(v) \tag{6}$$

These moving curves of discontinuity in  $u$  will represent very steep traveling fronts, and so  $u$  plays the role of the propagator variable. The

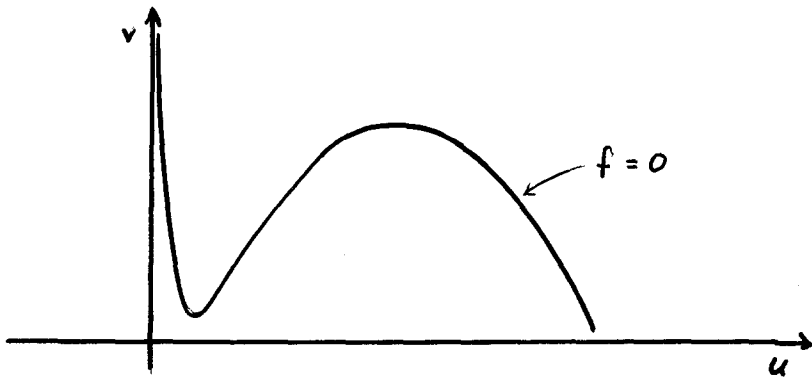


Fig. 1

fronts' velocities (and their fine-structure profile as well) are modulated by the controller variable  $v$  through the relation (6).

#### 4. SOLITARY PULSES

Of course, the dynamics of these pattern-forming fronts will depend very much on the nature of the function  $g$  appearing in (2) and (4). Suppose now that  $g = 0$  along a curve intersecting the  $f$ -nullcline as shown in Fig. 2, and that  $g$  has the signs shown in that same figure. Then the point  $(u_0, v_0)$  of intersection will be a stable rest state.

Consider a one-dimensional medium of infinite extent, and impose initial data  $u(x, 0), v(x, 0)$  such that  $u = u_0$  and  $v = v_0$  everywhere except on a finite interval of the  $x$  axis. Suppose also that in this finite interval, this pair of initial data traces a curve in the  $u-v$  plane (with  $x$  as parameter on the curve) which makes an excursion past the middle branch, as depicted in Fig. 2 (curved dotted line). Then on a time scale  $O(\epsilon)$ , the two branches  $h_{\pm}(v)$  will attract the portions of the initial curve which are nearest to them, and two steep fronts will form at the two values of  $v$  corresponding to the two points where the initial curve crosses the middle branch of the nullcline. These fronts will move according to their intrinsic trigger velocity (6), and moreover the one with the larger value of  $v$  may become transformed into a phase front,<sup>(9,25,34)</sup> when the value of  $v$  on the front attains the upper limit  $\bar{v}$ . This process of formation is also explained in Ref. 8; similar processes were described in Refs. 27 and 28.

As time passes, the two fronts will attain approximately the same velocity, either to the right or to the left, and the total configuration will resemble a somewhat flat-topped pulse. The leading front in this pulse will represent a transition from the minus (-) to the plus (+) state when

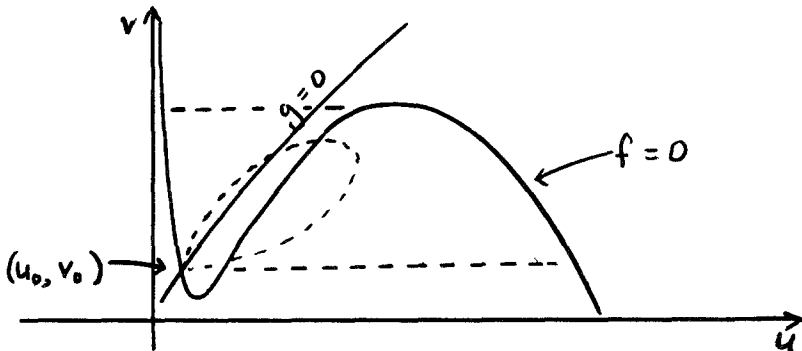


Fig. 2

$v = v_0$ . This transition is depicted by the lower horizontal dotted line in Fig. 2. The trailing front, which could be called a "back",<sup>(34)</sup> is really a "phase" front because it does not move with trigger velocity  $c(v)$ , but rather its trajectory is determined as the curve in the  $x-t$  plane where  $v(x, t) = \bar{v}$ . It is depicted by the upper dotted line in Fig. 2.

## 5. RING PATTERNS

Two-dimensional analogs of the solitary pulses described above exist in the form of straight bands propagating in one direction (normal to the band) in space (really a one-dimensional phenomenon) or solitary circular bands propagating outward. As long as their curvature is small compared with  $1/\varepsilon$ , the latter's motion along a given ray from the origin can be visualized and analyzed, to lowest order, the same way as the straight bands. Furthermore, if there is some mechanism at one point in this medium for the periodic generation of new bands, then there may result an unending succession of them. Such a mechanism apparently occurs in the BZ reagent, because the expanding ring (target) patterns commonly seen therein fit this description.

Two general mechanisms for the constant production of waves at the center have been proposed. The first, which undoubtedly occurs predominantly if not always, is the action of some inhomogeneity at the center causing the chemistry to change in a neighborhood of that point. This could be reflected in a change in the rate constants of the various reactions in the Oregonator model, so that they could in fact depend on position, assuming different values near the center from their values far away from it. Such a change in the rate constants could very well cause a shift in the relative positions of the  $f$  and the  $g$  nullclines from the configuration in Fig. 2 that in Fig. 3. In the latter, the intersection is on the unstable middle branch, and the diffusionless equations (1) and (2) with the Laplacian terms on the right omitted have solutions of relaxation oscillator type.

In this scenario, the medium could therefore be characterized as being oscillatory near the center, and as possessing a stable (but excitable) rest state outside of that central region. It is intuitively suggestive that such an inhomogeneous center may result in the production of a series of expanding rings, and in fact this production mechanism is verified and examined in great detail in Ref. 34.

The other type of mechanism proposed for the appearance of target patterns does not involve the presence of external inhomogeneities, but rather relies on the intrinsic properties of the reaction mechanism and the

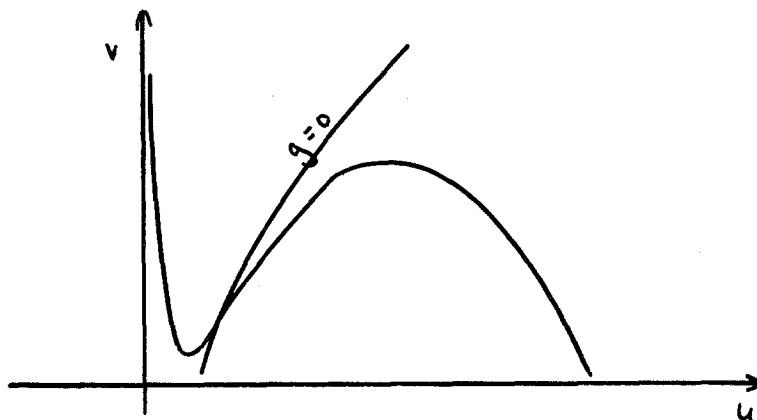


Fig. 3

diffusion of the species. For this to be possible, three equations rather than two are needed. One such system of three equations, not derived from any known chemical model, was suggested by Zaikin and Kawczynski,<sup>(40)</sup> and some numerical computations were done using that model. Models capable of similar effects were suggested in later papers by one of the those same authors.<sup>(17,18)</sup> In Ref. 5 I presented a model in which the reduced Oregonator PC system used in Ref. 34 was supplemented by a third very simple hypothetical equation involving slow reactions and a third chemical species. It was shown by asymptotic analysis, together with a stability argument that such a system was capable of producing inhomogeneous-center patterns. (Of course there has to be some inhomogeneity in the initial concentrations of the reactants for leading centers to arise.)

## 6. THE BIRTH OF SPIRALS

Some thought has been given to the process by which spirals can arise in an excitable medium. For example, a simple distributed dynamical system with three states at each (discrete) point in space-time was proposed and considered in Ref. 13. A close examination of this system elucidates to some extent the reason for spirals forming in that context.

In the context of PC models, such as (1), (2), a thought experiment was suggested in Ref. 9. The thought experiment involves a propagating straight (or circular) band pulse in a two-dimensional medium, as described in Section 4. If such a band is disturbed by, for example, physically mixing the chemicals in some region overlapping with a finite

segment of the band, then the pulselike structure is momentarily disturbed in that region. However, very soon at each point of the disturbed region the state vector  $(u, v)$  will be attracted to whichever branch of (5) it happens to be in the basin of attraction of. So in fact soon after the mixing, space will again be partitioned into two regions  $\Omega_{\pm}$ . Outside the disturbed region, of course,  $\Omega_{+}$  will simply be the flat-topped interior of the traveling pulse and  $\Omega_{-}$  will be everything else. It may well happen that the new configuration will be such that  $\Omega_{+}$  is no longer connected, but rather something as in Fig. 4. We assume that it has two connected components as shown.

Given such a readjusted configuration, one can now trace the movement of the boundaries between the two regions by using the law (6) to govern their normal velocity at each point, together with (4) and (5) to determine the evolution of the controller variable  $v$ . Most of the following aspects of this subsequent evolution process were brought out in Ref. 9.

Since  $v = v_0$  on the forward side of the moving pulse and  $v = \bar{v}$  on the aft side, and  $v$  is continuous along the boundary of each component, there will be a point  $P$  on each boundary at which  $v = v^*$ , where  $v^*$  is the point at which  $c(v^*) = 0$ . It is characterized by the "equal-area" rule:  $\int_{h_{-}(v^*)}^{h_{+}(v^*)} f(u, v^*) du = 0$ . Initially, this point serves as a pivot point about which the interface rotates as shown in Fig. 4. This rotation causes an asymmetry in what may well have been a symmetric initial configuration as in that figure.

The consequences of continuing with the reduced model (4)–(6) will now be described. Because the portion of the interface segment near  $P$  where  $v > v^*$  rotates into  $\Omega_{+}$ , where  $g > 0$ , it follows from (4) that  $v$  increases at a rate which is  $O(1)$  on that portion. This increase in  $v$  induces, through (6), a corresponding increase in the normal velocity of that segment. This means that arbitrarily close to  $P$ , the interface will soon develop a velocity which is bounded away from zero, as well as values of  $v$  bounded away from  $v^*$ . The same is true on the portion of the interface near  $P$  where  $v < 0$ , but the normal velocity in that case is in the opposite direction (though the angular velocity is in the same direction). Therefore

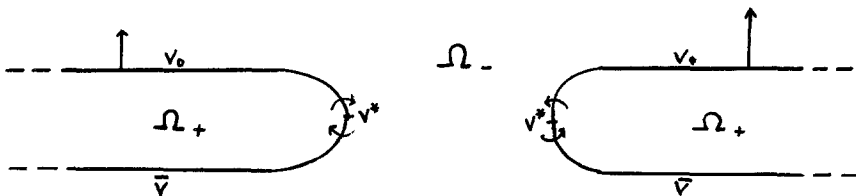


Fig. 4



the angular velocity of this segment near  $P$  becomes unbounded, and  $v$  becomes discontinuous. Moreover, the curvature of the interface also becomes unbounded. This violates the original hypothesis, used in forming the model, that the curvature is small compared to  $1/\epsilon$ . That reduction therefore may no longer be used near the pivot point  $P$ . The fact is that part of the reduction was to neglect the diffusion term in (2), supposing that there is no mechanism for the function  $v$  to develop large gradients. Such a mechanism has now appeared, and any further analysis near  $P$  must take that term into consideration; it will have the effect of smoothing out the discontinuity in  $v$  tempering the unbounded growth in angular velocity.

Irrespective of the analysis near  $P$ , which has not been done and would require some kind of rescaling, the quality of the motion of the interface in regions away from  $P$  can be gleaned by continuing the thought experiment. The pivot point  $P$  remains more or less stationary, the interface more or less rotating around it, while far away from it, the semiinfinite strip which is  $\Omega_+$  continues to act like a pulse, propagating upwards at constant velocity.

The effect of these two dynamical properties is to create rotating centers, which are more or less fixed, connected to unidirectional pulses. The rotating center winds the region  $\Omega_+$  into spiral-shaped strips which gain one more loop after each rotation (Fig. 5).

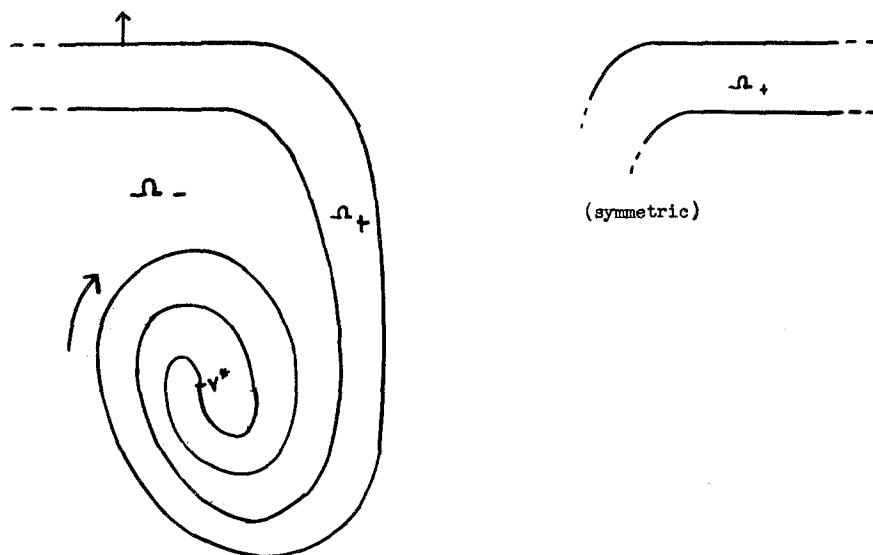


Fig. 5

Locally, the spiral arm looks like a relatively (compared to the undisturbed pulse) thin pulse propagating outward. Eventually, the spiral interface between  $\Omega_+$  and  $\Omega_-$  will be very long. On that interface, the function  $v$  will still vary continuously from the value  $\bar{v}$  to  $v_0$ , as it did originally. This means that along most of this boundary,  $v$  will have a very small gradient in the tangential direction.

## 7. DEVELOPED SPIRALS

Now let us suppose that a long time has elapsed, so that the spiral structure has developed a large number of loops and is rotating at a constant angular velocity. I shall try to show that this developed spiral cannot have uniform characteristic spatial dimensions (say, the typical width of the spiraling band), unless this characteristic dimension is smaller than that of the original pulse.

Suppose that the characteristic length were  $O(1)$ , so that the curvature of the interface is also uniformly  $O(1)$ . Now fix attention on some point  $Q$  at a positive distance  $r$  from the center, and let the spiral rotate past that point. During one rotation of the spiral, the point  $Q$  will be in  $\Omega_+$  some amount of time  $T_+$ , and in  $\Omega_-$  the amount of time  $T_-$ . These two times add up to  $T$ , the period of the rotation, which of course is independent of  $r$ . The individual times may depend somewhat on  $r$ , but neither is very small relative to  $T$ .

It may be assumed that the function  $g$ , which does not depend on  $\varepsilon$ , is such that  $g(h_{\pm}(v), v)$  is bounded away from 0 for  $v$  on the interface, as long as  $v$  does not stray too far from the value  $v^*$ . This function is positive when the  $+$  sign is taken, and negative when the  $-$  sign is taken. In view of (4), one sees that the function  $v(Q, t)$  increases at a nonzero rate during the time interval  $T_+$ , and decreases at a nonzero rate during the interval  $T_-$ . Since these rates are bounded away from 0, it follows that the total variation of  $v$  during one period is  $O(T)$ . Its maximum and minimum are attained at the interface itself.

By (6), the normal velocity of the fronts will also be of this same order. But this velocity is, in order of magnitude, equal to the characteristic normal spacing between the loops [assumed to be  $O(1)$ ] divided by the characteristic time  $T$ , so one obtains  $T = O(1/T)$ , or  $T = O(1)$ . Hence the value of  $v$  on the interface is such that  $v - v^* = O(1)$ , uniformly in  $r$ , and  $v$  cannot smoothly approach the value  $v^*$  at the center.

It follows either that the developed spiral must be described by at least two different space scales, or by a uniform scale not equal to  $O(1)$ . In the latter case, the scale could not be larger than that of the solitary pulse,

which is  $O(1)$ , so the scale would have to be  $\ll 1$ . Exploring the possibility of a small uniform spatial scale, one finds that scaling space by the factor  $\delta^2 \equiv \varepsilon^{2/3}$  leads to a reasonable problem for the fully developed spiral, provided that time is scaled by the factor  $\delta$ .<sup>(10)</sup> I shall outline how the argument proceeds.

A free boundary problem is found for the determination of the shape of the interface, in these scaled coordinates, and the function  $v$ . It happens that  $v$  differs from  $v^*$  by a small amount of order  $\delta \equiv \varepsilon^{1/3}$ . In that range, the function  $g(h_{\pm}(v), v)$  is approximately constant, of the same sign as the subscript of  $h$ , so for simplicity this function can be taken to be  $\pm 1$ . It is also convenient to reposition the origin on the  $v$  axis to be at  $v^*$ , so replace  $v$  by  $v^* + v$ . Finally, simplicity again suggests making  $f$  symmetric:  $f(-u, -v) = -f(u, v)$ . [It follows from this that  $h_-(0) = -h_+(0)$  (Fig. 6).]

Now make the scale changes

$$\xi \equiv \delta^{-2}x, \quad \tilde{v} \equiv \delta^{-1}v$$

In terms of the polar coordinates  $(r, \tilde{\theta})$  in the  $\xi$  plane and angular velocity  $\delta^{-1}\omega$ , the rotating solution will be represented by functions

$$u = u(r, \tilde{\theta} - \delta^{-1}\omega t), \quad \tilde{v} = \tilde{v} \text{ (same)}$$

the basic equations (1) and (2), under these variable changes, become

$$f(u, \delta\tilde{v}) + \delta^2(\Delta_{\xi}u + \omega u_{\tilde{\theta}}) = 0 \tag{7}$$

$$\Delta\tilde{v} + \omega\tilde{v}_{\tilde{\theta}} + g(u, \delta\tilde{v}) = 0 \tag{8}$$

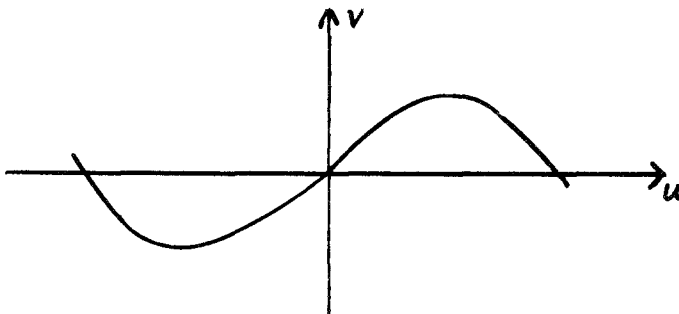


Fig. 6

where  $\Delta$  is the Laplacian in the coordinates  $\xi$ . Let us agree to drop the tildes from the symbol  $\tilde{v}$ .

To lowest order in the small parameter  $\delta$ , (7) gives  $f(u, 0) = 0$ , which means that  $u = h_{\pm}(0)$  (recall that now  $v^* = 0$ ). Therefore  $g = \pm 1$  in (8), and one obtains

$$\Delta v + \omega v_{\xi} \pm 1 = 0 \tag{9}$$

for  $\xi \in \Omega_{\pm}$ .

One seeks solutions such that the interface  $\Gamma$  between  $\Omega_+$  and  $\Omega_-$  is spiral shaped, and is given in two parts:  $\Gamma_+$ , described by  $\tilde{\theta} = h(r)$  for some function  $h$ , where  $r = |\xi|$ , and  $\Gamma_- : \tilde{\theta} = h(r) + \pi$ . As  $r \rightarrow \infty$ , we require that

$$h(r) = \gamma r + O(1) \tag{10}$$

which guarantees that the spiral is Archimedian, the type usually seen in experiments. Here  $\gamma > 0$  is the unknown pitch of the spiral far from the origin.

The problem is most naturally written in terms of the shifted polar coordinates  $(r, \theta)$ , where

$$\theta \equiv \tilde{\theta} - h(r)$$

Defining the function  $H(r) \equiv h'(r)/r$ , one finds that the Laplacian term assumes the form

$$\Delta_H u \equiv \frac{1}{r} (ru_r)_r + \frac{1}{r^2} u_{\theta\theta} - \frac{1}{r} (r^2 H)' u_{\theta} - 2rHu_{r\theta} + r^2 H^2 u_{\theta\theta} \tag{11}$$

Along with the equation (9), which must hold in the two domains  $\Omega_{\pm}$ , an interface condition on the boundary  $\Gamma$  (characterized by  $\theta = 0$  or  $\theta = \pi$ ) separating the two domains must be imposed, in order to have a reasonable problem. The correct interface condition can be found by returning to the original equation (1) for  $u$ , and using it to examine the fine structure of the transition that  $u$  undergoes on the boundary  $\Gamma$ . This fine structure analysis is accomplished by stretching the angular variable  $\theta$  near  $\Gamma$ , i.e., near  $\theta = 0$ . Thus, define

$$\psi \equiv \frac{\theta}{\delta}, \quad U(r, \psi) \equiv u(r, \theta), \quad V(r, \psi) \equiv v(r, \theta)$$

With these variables, (1) becomes

$$\begin{aligned} \frac{1}{r^2} U_{\psi\psi} + r^2 H^2 U_{\psi\psi} + f(U, \delta V) - \frac{\delta}{r} (r^2 H)' U_{\psi} \\ - 2\delta r H U_{r\psi} + \delta \omega U_{\psi} + \frac{\delta^2}{r} (rU_r)_r = 0 \end{aligned} \tag{12}$$

The location of the interface is defined, in the stretched coordinates, as the place where  $U=0$ , and the angular variable is adjusted to vanish there:

$$U(r, 0) = 0$$

Expand the solution formally in a series  $U = U^0 + \delta U^1 + \dots$ ,  $V = V^0 + \dots$ , and substitute into (12). To lowest order, one obtains

$$\left(\frac{1}{r^2} + r^2 H^2\right) U_{\psi\psi}^0 + f(U^0, 0) = 0, \quad U^0(r, 0) = 0$$

The solution which matches with the outer solution  $u = h_{\pm}(0)$  as  $\psi \rightarrow \pm\infty$  is

$$U^0(r, \psi) = \chi(p(r) \psi)$$

where  $\chi(s)$  is the well-known stationary front solution of the problem

$$\chi'' + f(\chi, 0) = 0, \quad \chi(0) = 0, \quad \chi(\pm\infty) = h_{\pm}(0)$$

and  $p(r) \equiv [r^{-2} + r^2 H(r)^2]^{-1/2}$ . Carrying out the solution to the next order in  $\delta$  involves applying a solvability condition, because the linearized operator

$$L = \frac{d^2}{ds^2} + f_u(\chi(s), 0)$$

with zero boundary conditions at  $\pm\infty$ , has a null space spanned by the single function  $\chi'(s)$ . This next order problem's solvability condition then provides<sup>(10)</sup> the appropriate interface condition, which is

$$v(r, 0) = M \left( \left\{ \frac{1}{r} [r^2 H(r)]' - \omega \right\} p(r) + r H(r) p'(r) \right) \quad (13)$$

$M$  being a specific constant.

Finally, the symmetry condition imposed upon  $f$  induces a corresponding symmetry condition on the solution:  $v(r, -\theta) = -v(r, \theta)$ .

The problem for the spiral can now be stated: Find a function  $v(r, \theta)$ , a function  $H(r)$ , and constants  $\omega$  and  $\gamma$ , satisfying

$$(1) \quad \Delta_H v + \omega v_0 \pm 1 = 0$$

The + sign holding for  $\theta \in (0, \pi)$  and the - sign for  $\theta \in (-\pi, 0)$ ,

$$(2) \quad v(r, \theta + \pi) = -v(r, \theta)$$

$$(3) \quad H(r) = \frac{\gamma}{r} + O(r^{-2}) \quad (r \rightarrow \infty)$$

and (4) Eq. (13).

An asymptotic analysis of this problem for large distances  $r$  from the center provides a solution there for every value of  $\omega$  in some interval  $(0, \omega_0]$ . Formal solutions for small values of  $r$  can also be constructed in the form of power series in  $r$ .

More mathematical and numerical work needs to be done to answer the questions raised by this problem. In particular, an existence theorem needs to be provided for it, and a numerical method needs to be devised for solving it. It may happen that a solution exists for a range of values of  $\omega$ , as is suggested by the large- $r$  expansion. In that case, a method needs to be found for deciding which value of  $\omega$  is the preferred one, as experiments seem to indicate that a unique spiral, with a unique value of  $\omega$ , is associated with each given reagent.

## 8. CONCLUSION

It has been pretty well established that the PC system examined in this talk is a realistic model for the spatially distributed BZ reagent under commonly occurring parameter conditions. The analysis of the system is straightforward and provides a clear picture of the mechanism behind the appearance of fronts, trains, solitary pulses, and target patterns, and behind the generation of spirals, under those conditions. In fact, the PC model has been remarkable 'successful in reflecting observed phenomena (see a discussion of this point in Ref. 34). Spiral-like structures in 3-space have been called scrolls by A. Winfree; their generation can also be easily understood along these same lines, as resulting from local disturbances of plane solitary waves propagating through 3-space. Fully developed spirals can likely be understood in this same PC context as solutions of the free boundary problem presented in Section 7; more mathematical and numerical work should be done on that problem.

Very possibly other parameter conditions exist for which similar spatial patterns can be seen in the BZ reagent, but for which the PC

framework is not appropriate; and very likely other excitable biological or chemical media, supporting such patterns, may require other models for their understanding. Nevertheless, it appears likely that models having many of the properties of the one studied here will play a significant role in the unraveling of these more complex systems.

## ACKNOWLEDGMENT

This research was supported in part by NSF grant No. DMS-8202056.

## REFERENCES

1. D. S. Cohen, J. C. Neu, and R. R. Rosales, Rotating spiral wave solutions of reaction-diffusion equations, *SIAM J. Appl. Math.* **35**:536 (1978).
2. R. J. Field, E. Korös, and R. M. Noyes, Oscillations in chemical systems. II. Thorough analysis of temporal oscillation in the bromate-cerium-malonic acid system, *J. Am. Chem. Soc.* **94**:8649-8664 (1972).
3. R. J. Field and R. M. Noyes, Explanation of spatial band propagation in the Belousov reaction, *Nature* **237**:390-392 (1972).
4. R. J. Field and R. M. Noyes, Oscillations in chemical systems. IV. Limit cycle behavior in a model of a real chemical reaction. *J. Chem. Phys.* **60**:1877-1884 (1974).
5. P. C. Fife, On the question of the existence and nature of homogeneous-center target patterns in the Belousov-Zhabotinskii reagent, in *Analytical and Numerical Approaches to Asymptotic Problems in Analysis*, O. Axelsson *et al.*, eds., Mathematics Studies No. 47 (North-Holland, Amsterdam, (1981), pp. 45-56.
6. P. C. Fife, *Mathematical Aspects of Reacting and Diffusing Systems*, Lecture Notes in Biomathematics No. 28 (Springer-Verlag, New York, 1979).
7. P. C. Fife, Pattern formation in reacting and diffusing systems, *J. Chem. Phys.* **64**:854-864 (1976).
8. P. C. Fife, Sigmoidal systems and layer analysis, in *Competition and Cooperation in Neural Nets*, S. Amari and M. A. Arbib eds., Lecture Notes in Biomathematics No. 45 (Springer-Verlag, Berlin, 1982), pp. 29-56.
9. P. C. Fife, Current topics in reaction-diffusion systems, in *Nonequilibrium Cooperative Phenomena in Physics and Related Fields*, M. G. Velarde, ed. (Plenum Press, New York, 1984), pp. 371-412.
10. P. C. Fife, Propagator-controller systems and chemical patterns, in *Non-equilibrium Dynamics in Chemical Systems*, A. Pacault and C. Vidal, eds. (Springer-Verlag, Berlin, 1984).
11. J. Greenberg, Axisymmetric time-periodic solution of reaction-diffusion equations, *SIAM J. Appl. Math.* **34**:391-397 (1978).
12. J. Greenberg, Spiral waves for  $\lambda$ - $\omega$  systems, *SIAM J. Appl. Math.* **39**:301-309 (1980).
13. J. M. Greenberg, B. D. Hassard, and S. P. Hastings, Pattern formation and periodic structures in systems modeled by reaction-diffusion equations, *Bull. Am. Math. Soc.* **84**:1296-1327 (1978).
14. P. Hagan, Spiral waves in reaction-diffusion equations, *SIAM J. Appl. Math.* **42**:762-785 (1982).
15. L. N. Howard and N. Kopell, Slowly varying waves and shock structures in reaction-diffusion equations, *Stud. Appl. Math.* **56**:95-145 (1977).

16. G. R. Ivanitskii, V. I. Krinsky, and E. E. Sel'kov, *Mathematical Biophysics of a Cell* (Nauka, Moscow, 1978).
17. A. L. Kawczynski and J. Gorski, A model of nonsustained leading center, *Bull. l'Acad. Pol. Sci.* **24**:261–268 (1981).
18. A. L. Kawczynski and J. Gorski, Chemical models of non-sustained leading center, *Pol. J. Chem.* (to appear).
19. J. P. Keener, Waves in excitable media, *SIAM J. Appl. Math.* **39**:528–548 (1980).
20. N. Kopell, Target pattern solutions to reaction–diffusion equations in the presence of impurities, *Adv. Appl. Math.* **2**:389–399 (1981).
21. N. Kopell, Forced and coupled oscillators in biological applications, in *Proceedings of the International Congress of Mathematics, Warsaw, 1983*.
22. N. Kopell and L. N. Howard. Target patterns and spiral solutions to reaction–diffusion equations with more than one space dimension, *Adv. Appl. Math.* **2**:417–449 (1981).
23. Y. Kuramoto, *Chemical Oscillations, Waves, and Turbulence*, Springer Series in Synergetics No. 19 (Springer-Verlag, New York, 1984).
24. A. S. Mikhailov and V. I. Krinsky, Rotating spiral waves in excitable media: the analytical results, *Physica* **9D**:346–371 (1983).
25. P. Ortoleva and J. Ross, Phase waves in oscillatory chemical reactions, *J. Chem. Phys.* **58**:5673–5680 (1973).
26. P. Ortoleva and J. Ross, On a variety of wave phenomena in chemical oscillations, *J. Chem. Phys.* **60**:5090–5107 (1974).
27. P. Ortoleva and J. Ross, Theory of propagation of discontinuities in kinetic systems with multiple time scales: fronts, front multiplicity, and pulses, *J. Chem. Phys.* **63**:3398–3408 (1975).
28. L. A. Ostrovskii and V. G. Yahno, The formation of pulses in an excitable medium, *Biofizika* **20**:489–493 (1975).
29. Z. Noszticzus, H. Farkas, and Z. A. Schelly, Expodator: a new skeleton mechanism for the halate driven chemical oscillators, *J. Chem. Phys.* **80**:6062–6070 (1984).
30. R. M. Noyes, An alternative to the stoichiometric factor in the Oregonator model, *J. Chem. Phys.* **80**:6071–6078 (1984).
31. J. Tyson, *The Belousov–Zhabotinskii Reaction*, Lecture Notes in Biomathematics No. 10 (Springer-Verlag, Berlin, 1976).
32. J. Tyson, Oscillations, bistability, and echo waves in models of the Belousov–Zhabotinskii reaction, *Ann. N. Y. Acad. Sci.* **36**:279–295 (1979).
33. J. Tyson, On scaling and reducing the Field–Körös–Noyes mechanism of the Belousov–Zhabotinskii reaction, *J. Phys. Chem.* **86**:3006–3012 (1982).
34. J. Tyson and P. C. Fife, Target patterns in a realistic model of the Belousov–Zhabotinskii reaction, *J. Chem. Phys.* **73**:2224–2237 (1980).
35. A. T. Winfree, Spiral waves of chemical activity, *Science* **175**:634–636 (1972).
36. A. T. Winfree, Wavelike activity in biological and chemical media, in *Lecture Notes in Biomathematics*, P. van den Driessche, ed. (Springer-Verlag, Berlin, 1974).
37. A. T. Winfree, Rotating solutions of reaction–diffusion equations in simply-connected media, in *SIAM–AMS Proceedings*, Vol. 8, Providence (1974), Pp. 13–31.
38. A. T. Winfree, Stably rotating patterns of reaction and diffusion, in *Theoretical Chemistry*, Vol. 4, H. Eyring and D. Henderson, eds. (Academic Press, New York, 1978), pp. 1–51.
39. A. T. Winfree, The rotor as a phase singularity of reaction–diffusion problems and its possible role in sudden cardiac death, in *Nonlinear Phenomena in Chemical Dynamics*, C. Vidal and A. Pacault, eds. (Springer-Verlag, Berlin, 1981), pp. 156–159.
40. A. N. Zaikin and A. L. Kawczynski, Spatial effects in active chemical systems. I. Model of leading center, *J. Non-Equilib. Thermodyn.* **2**:39–48 (1977).



41. A. M. Zhabotinskii, *Concentrational Self-Oscillations* (Nauka, Moscow, 1974).
42. A. M. Zhabotinsky and A. N. Zaikin, Concentration wave propagation in two dimensional liquid-phase self-oscillating systems, *Nature (London)* **225**:535–537 (1970).
43. A. M. Zhabotinsky and A. N. Zaikin, Auto-wave processes in a distributed chemical system, *J. Theor. Biol.* **40**:45–61 (1973).