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PROPAGATING WAVES AND TARGET PATTERNS IN CHEMICAL SYSTEMS.(U)
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MRC Technical Summary Report #2074

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May 1980

Received December 14, 1979

Sponsored by

U. S. Army Research Office
P. O. Box 12211
Research Triangle Park
North Carolina 27709

National Science Foundation
Washington, D. C. 20550

UNIVERSITY OF WISCONSIN - MADISON
MATHEMATICS RESEARCH CENTER

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ABSTRACT

A class of models for target patterns (concentric circular waves emanating from a point called the leading center) is constructed in the context of singularly perturbed reaction-diffusion systems of partial differential equations. First, the theory of wave fronts is detailed for scalar equations and systems of equations. A scaling method reduces complex waves to the consideration of a group of simple wave phenomena. It is shown that expanding wave fronts can be generated spontaneously at a point. This process, together with the laws of their subsequent motion, reduces the problem to an ordinary differential initial value problem, whose solution is required to have certain properties. A discussion is given of the connection between these results and experimental observations.

AMS(MOS) Subject Classification: 35K55, 80A20, 35B25

Key words: Reaction, Diffusion, Chemical waves, Wave front, Pattern formation, Singular perturbations

Work Unit No. 2 - Physical Mathematics

*This is a revised and expanded version of a paper presented by Fife at a conference with proceedings published as Dynamics of Synergetic Systems, H. Haken, ed, Springer-Verl (1980).

Sponsored by the United States Army under Contract Nos. DAAG29-75-C-0024 and DAAG29-80-C-0041 and National Science Foundation Grant No. MCS79-04443.

SIGNIFICANCE AND EXPLANATION

Propagating chemical waves, particularly those emanating from a "leading center" or those forming a spiral, may be seen in laboratory reagents involving the Belousov-Žhabotinskiǐ reaction. Chemical and physico-chemical waves also occur in biological media. Typically, these phenomena exhibit multiple natural time and space scales. The mathematical treatment of these waves consists in setting up an appropriate model for them, and then analyzing it. A natural type of model in the present situation involves a system of partial differential equations of reaction-diffusion type; small parameters may be put into the system to effectuate the multiple scales. This paper explains the basic steps in modeling chemical waves this way, and applies them to the case of target patterns (concentric circular waves emanating from a point). The models constructed here reflect the known qualitative kinetics of the BZ reaction. The techniques are expected to be of value in reaction-diffusion-convection problems as well.

The responsibility for the wording and views expressed in this descriptive summary lies with MRC, and not with the authors of this report.

PROPAGATING WAVES AND TARGET PATTERNS IN CHEMICAL SYSTEMS*

Paul C. Fife and John Tyson

1. Introduction.

The discovery of propagating waves of various types in chemical reagents has provoked a great deal of research, during the last ten years, into the phenomenology and the underlying mechanisms for such wavelike activity. The research has been performed by natural scientists and mathematicians alike. Most of it has been experimental, but much computer simulation and mathematical analysis has also been done. Chemical wave activity is believed to be prevalent in biological organisms, but the most readily accessible reagent for laboratory study is that discovered by Belousov and Zabotinskii (the Z-reagent). This mixture has oscillatory or excitable kinetics, depending on the concentrations of the various chemicals in the solution. Both of these regimes have at least two natural time scales: During one period of an oscillation or during one excited "excursion", most of the variation in the concentration of the reactants occurs within a brief interval of time. The time scale associated with this brief spurt of activity is much shorter than that associated with the slow variation which occurs before and after. This is well known from experiment and computation, and is evident from scaling analyses of model kinetic equations performed in [1] and elsewhere. Spatial structures are also prevalent in unstirred layers of this reagent ([23];[2], [22], [24], and references therein). Target patterns (expanding concentric circular waves) are among the most prevalent of these structures. Here again, disparate space and time scales are evident from computer simulation of propagating waves [3].

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Sponsored by the United States Army under Contract Nos. DAAG29-75-C-0024 and DAAG29-80-C-0041 and National Science Foundation Grant No. MCS79-04413.

It is natural, therefore, to use multiple scaling techniques when attempting to reduce wave and pattern phenomena to mathematical analysis. The study of propagation phenomena in excitable media such as biological membranes has indeed profited from this use [1-8]. In general reaction-diffusion settings, these methods were pursued in some detail in [9,10,26]. Nevertheless, their full implications have yet to be determined. In particular, their use in analyzing target patterns has been neglected. The purpose of the present paper is to explain some basic principles in the analysis of wave fronts by scaling techniques, and to discuss the application of these principles to the task of modeling target patterns.

We begin with a brief account, in Section 2, of the fundamental theory of wave fronts for scalar nonlinear diffusion equations, as these are the component parts of the more complex wave phenomena to be examined in later sections. A scaling method to reduce the study of more complex propagating fronts to that of scalar fronts is elaborated in Section 3. In 1974, Winfree [11] suggested that at least two broad categories of chemical waves exist: phase and trigger waves (see also [3]). We can see such a distinction more clearly in the context of wave fronts associated with relaxation oscillatory, (or even excitable) kinetics; this distinction is explained and discussed at some length in Section 4.

Besides treating the existence and properties of chemical structures, one may wish to inquire how they might arise in a reagent in the first place. Two such mechanisms are detailed in Section 5. Section 6 takes up the problem of modeling target patterns. It is found that the theory of sharp wave fronts, as developed in the preceding section, is very useful here. These patterns [illegible] from a series of [illegible] fronts, at least some of trigger type, generated periodically at one point. At first glance, the idea is conceptually clear; [illegible] difficulties arise when a mathematical analysis is performed. [illegible] oscillators, [illegible] Section 7. [illegible] [illegible] [illegible]

Although patterns in the Z-reagent are the motivation for this work, specifics of the Z-reagent chemistry are not touched upon here. We have made a detailed study in [27] of the patterned solutions of a system of equations which realistically models the chemistry of that reagent.

Other writers ([12,13], and especially Kopell and Howard [14]) have studied target patterns within the context of $\lambda-\omega$ systems. An approach via Padé approximants is in [25]. These approaches are entirely different from that discussed here. The reader interested in the Z-reagent can profit from the books by Zabotinskii [15] and by Tyson [16]. Some of the material in this paper was presented from a different point of view, and in more mathematical detail, in [17]. Some of the results announced here represent joint work with R. Smock. We wish to thank M. Marek for bringing to our attention papers [4] and [7].

2. Scalar fronts.

Here we review some basic facts about wave front solutions $u = U(x-ct)$ of scalar nonlinear diffusion equations

$$u_t = u_{xx} + f(u) \tag{1}$$

where f has two zeros: $f(U_1) = f(U_2) = 0$. Under fairly general circumstances, there exist fronts satisfying

$$U(-\infty) = U_1,\ U(\infty) = U_2. \tag{2}$$

Two important cases arise in applications:

(a) $f'(U_1) < 0$, $f'(U_2) < 0$, , and f has only one intermediate zero between U_1 and U_2;

(b) $f(u) \neq 0$ for u in the interval between U_1 and U_2.

In case (a), there exists a unique velocity c and a profile $U(z)$, unique up to shifts in z, satisfying (2), such that $U(x-ct)$ satisfies (1) [18]. This front is very stable; if it is perturbed by any bounded function whose bound does not surpass a certain known constant, then the resulting solution of (1) evolves, as $t \to \infty$, back to the same front (possibly shifted by a certain amount) [19].

In case (b), on the other hand, there exists a whole range of possible front velocities [20]. For example, suppose $f > 0$ and $U_1 > U_2$. Then there is a positive minimal speed c^* such that for any value c, $c^* \leq c < \infty$, there exists a unique (except for shifts) wave front solution $U(z)$, satisfying (2). These fronts are stable to small perturbations which are zero except in a finite interval, but they are certainly not stable to the same extent as those of type (a).

In any case, the front moves in such a direction that for each x, $u(x,t) = U(x-ct)$ approaches the "dominant state" as $t \to \infty$. The dominant state is defined to be the constant U_1 if $\int_{U_1}^{U_2} f(s)ds < 0$, and U_2 if this integral > 0. If the integral equals zero, then $c = 0$ and the front is stationary.

3. Decoupling and free boundary problems.

In typical singular perturbation problems, a complex system may be reduced to several simpler ones by rescaling and exploiting the smallness of some parameter. The simpler problems may govern the solution in different parts of its domain of definition; thus there may be boundary layers versus regions of relatively slow variation.

Analogous situations arise in reaction-diffusion problems [9,10]. To illustrate this, we consider a system of $n = n_1 + n_2$ reacting and diffusing components, n_1 of them "fast" and the others not fast. Let $u = (u_1,\dots,u_{n_1})$ be the vector of fast components, and v the others. The system of RD equations in one space variable is of the form

$$u_t = \alpha D_1 u_{xx} + kf(u,v), \tag{3a}$$

$$v_t = D_2 v_{xx} + g(u,v). \tag{3b}$$

Here D_i are (diffusion) matrices; $k >> 1$ is a parameter expressing the fact that reactions affecting the concentration u are fast; and the parameter $\alpha << k$ is inserted to account for the possibility that the diffusion rate of u may be small or large $(D_i = 0(1))$. The lowest order approximation, in regions where u_t and αu_{xx} are not large, is obtained by setting the coefficient of k equal to zero:

$$f(u,v) = 0. \tag{4}$$

We assume that this equation can be solved for u in a nonunique manner: there are at least two functions $h_+, h_- : R^{n_2} \to R^{n_1}$, such that (4) holds when

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

In other words, $f(h_+(v),v) \equiv f(h_-(v),v) \equiv 0$.

We imagine that the x-t plane is partitioned into two parts $\Omega_\pm$, in which (5) holds (approximately) with the corresponding sign. Under certain circumstances, such a partitioning is possible, with sharp wave fronts forming the boundaries between the two domains.

To investigate this further, we scale x and t differently in the layer between Ω_+ and Ω_-. The scaling will be chosen so as to eliminate the parameters in (3a).

Setting

$$\tau = kt, \quad \zeta = \sqrt{\frac{k}{\alpha}}\, x,$$

we reduce (3a) to

$$u_\tau = D_1 u_{\zeta\zeta} + f(u,v). \tag{6}$$

If we assume that $v(x,t)$ varies smoothly across the transition zone between Ω_+ and Ω_-, then within this narrow zone, v may be treated as constant. In this case it is reasonable to suppose that (6), like (1), has a traveling front solution connecting the two known zeros of f, namely $h_-(v)$ and $h_+(v)$ (if $n_1 = 1$, the theory is governed by the considerations in Section 2). Let us assume this is true, and that this front is a higher-dimensional analogue of the scalar front of type (a) in Section 2. That is, we assume the velocity c and profile are uniquely determined from the parameter v in (6). Thus $u = U(\zeta - c(v)\tau; v)$. This type of reasoning was used in [4], [9], and later papers.

In the original variables, $u = U(\sqrt{\frac{k}{\alpha}}\,(x - \sqrt{\alpha k}\, ct))$, revealing that the actual velocity is of the order $\sqrt{\alpha k}$, and the width of the front is of the order $\sqrt{\frac{\alpha}{k}} \ll 1$. Now let $x = y(t)$ denote the position of one such front. Knowing its velocity, we may write

$$\frac{dy}{dt} = \sqrt{\alpha k}\; c(v(y,t)). \tag{7}$$

Suppose there is only one such front, and Ω_- lies to the left of it, with Ω_+ to its right. Then to lowest order, we have found that when v is known, u is determined by (5) in $\Omega_\pm$, and that the boundary between Ω_+ and Ω_- moves according to (7). Thus, it appears that we have uncoupled u from v.

This is true in that the problem reduces to one for v alone; but it is a free boundary problem, [illegible] (3b) with the replacement (5) [illegible], the "+" [illegible] $x > y(t)$, the "−" [illegible], and [illegible] boundary $y(t)$ governed by a nonlinear [illegible] equation (7).

[illegible] negligible, [illegible]

In more general situations, the boundary between Ω_+ and Ω_- could consist of several wave fronts. And, of course, the extension of this reasoning to higher space dimensions is clear.

4 Phase and trigger fronts.

We refer to the setting in Section 3 with $n_1 = n_2 = 1$; corresponding phenomena in higher dimensions remain to be explored. So now u and v are scalar functions. The additional complication we impose is that $h_\pm$ are not defined for all values of v; rather their graphs lie on a nullcline of f as shown:

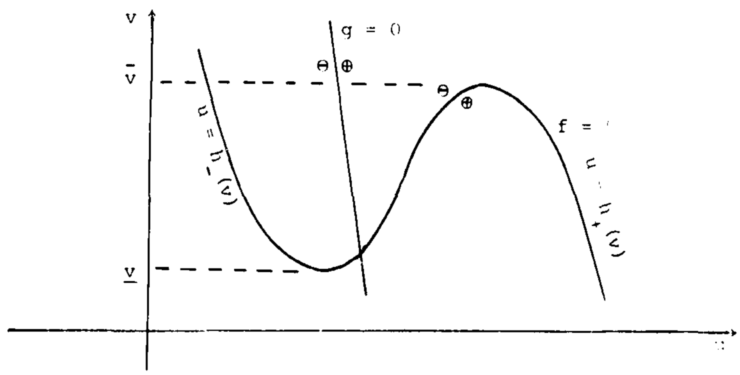

Figure 1

For values of v in the interval $\underline{v} < v < \bar{v}$, $f(u,v)$ has the features of the function in (1) in case (a), provided $f_u(h_\pm(v),v) \neq 0$.

We assume, mainly for simplicity, that $D_2 = 0$ in (3b), and that $g(h_-(v),v) \equiv G_-(v) < 0$, $g(h_+(v),v) \equiv G_2(v) > 0$. If there is a single front with trajectory $y(t)$ and v varying continuously across it, then

$$\frac{\partial v}{\partial t} = \begin{cases} G_-(v) < 0\ , & x < y(t)\ , \\ G_+(v) > 0\ , & x > y(t)\ . \end{cases} \qquad (\ )$$

If $\underline{v} < v < \bar{v}$, $y(t)$ is governed by (7), because the function $c(v)$ in that equation comes from consideration of the bistable case ((a), Section 2). Furthermore, if [illegible] initially continuous at the front (as we assume), it must remain so. For otherwise, [illegible] front passes a fixed value of x, v would change discontinuously in time, meaning that v_t would have a δ- function behavior. This is contradicted by (8): the right side, though discontinuous, is bounded.

Now suppose that $c = y' > 0$: the front is advancing into the region where $u \sim h_+(v)$. This will be the case when v is near $\underline{v}$, for then h_+ is the dominant state, according to the definition in Section 2. Then the values of $v(x,t)$ ahead of the front $(x > y)$ determine the motion of the front: $y' = c(v(y,t))$.

It may happen, however, that $v(y(t)t)$ attains the minimal value $\underline{v}$ at some time t_0. At that point, v is prohibited from any further decrease: there can exist no front with $v < \underline{v}$. We must therefore have, at $t = t_0$,

$0 = \frac{d}{dt} v(y(t),t) = v_x(y(t) + 0,t)y'(t) + v_t = v_x^+ y' + G_+(\underline{v})$, where v_x^+ is the x-derivative of v at the front evaluated from the right. Hence

$$\frac{dy}{dt} = -G_+(\underline{v})/v_x^+ . \tag{9}$$

This relation replaces (7) when v attains the value $\underline{v}$, in fact continues to hold as long as $v(y,t) = \underline{v}$.

We may inquire whether a front can exist when $v = \underline{v}$, for at this value of v, we are not in the bistable case (a). However, we are in case (b), as $f(u,\underline{v})$ is of one sign for $h_-(\underline{v}) < u < h_+(\underline{v})$, and fronts exist in this case as well. In fact, as we have seen, their velocity is arbitrary, subject only to a minimal value c^*. This means that one with velocity given by (9) is indeed possible.

We therefore have two types of propagation laws for fronts: (7) and (9). These types correspond to "trigger" and "phase" waves, in the terminology of Winfree [11]. To summarize, trigger fronts occur when $\underline{v} < v < \bar{v}$; their speed is determined by (7), where the function c comes from a law for scalar fronts, and is produced by the combination of diffusion and reaction. Phase fronts, on the other hand, occur when $v = \underline{v}$ or $\bar{v}$; their speeds are totally unaffected by either diffusion or the reaction term f. Rather (from (9)) they depend on the distribution of values of v ahead of the front (specifically, on v_x^+). In this sense, the motion of phase fronts is determined by the initial values of v.

5. The generation of fronts.

We ask now, by what process may fronts be formed, if they do not originally exist. There are two such processes:

(i) (See [9].) Referring back to Figure 1, suppose that, at time $t = 0$, the pair of functions $(u(x,0), v(x, $ [illegible] map the x-axis on to a curve Γ in the (u-v) plane as shown:

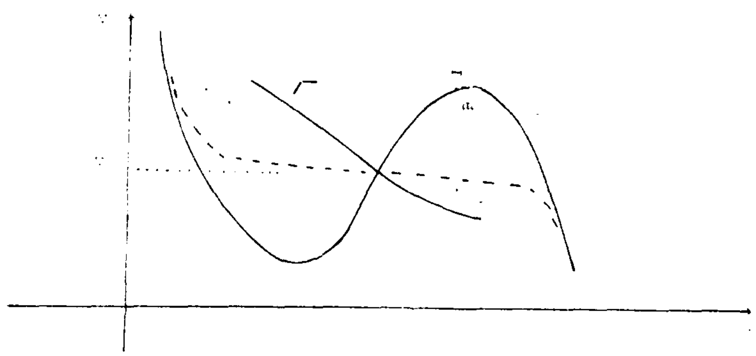

Figure 2

Initially, the second term in (3a) is negligible compared to the last term, and we have $u_t \sim kf(u,v)$. This means that as time increases, u rapidly (since $k >> 1$) changes in such a manner that the phase-plane image is drawn from its initial position Γ to one or both of the stable descending branches $u = h_\pm(v)$. If Γ intersects the ascending intermediate branch (as shown) at some value $v = v_0$, then the evolved image curve is split between the two stable branches, as shown by the dotted line. At some point the x-variation in u will be abrupt enough that the first term on the right of (3a) is no longer negligible; and at this point, a sharp wave front in u is formed, with v varying continuously across the front, attaining the value v_0 at the front itself.

(ii) (See [illegible].) Suppose, on the other hand, that Γ does not intersect the intermediate branch. Then the image is attracted entirely to only one of the stable branches, say h_-. After this happens, a slower process takes place, in which v evolves according to (3b) with u replaced by $h_-(v)$. Again for simplicity, assume $D_2 = 0$, so $v_t = G_-(v) < 0$. Eventually, for some $x = x_0$, v will attain a minimal value of $\underline{v}$. Further decrease of v causes the image to leave the branch $u = h_-(v)$ for x in a neighborhood of x_0. Then by the process described in (i) above, that part of the image curve is rapidly attracted to the other stable branch $u = h_+(v)$. This localized attraction to h_+ causes a pair of fronts, facing oppositely, to be formed near $x = x_0$. For each such front, h_+ will be the dominant state, so the fronts will move apart, increasing the interval on which $u \sim h_+(v)$.

6. Target patterns.

These are a series of concentric circular chemical waves, expanding outward, new ones regularly being generated at the center (usually called a "leading center" [21]). Such patterns have been observed in various forms of the Z-reagent ([23]; [2], [22], and references therein). Some of the patterns observed are associated with externally imposed heterogeneities at the center. We shall indicate in (i) below how such targets may be modelled. Self-sustaining target patterns, not dependent upon external stimuli, may also be modelled by the techniques discussed above; see (ii) below.

All of the models we describe involve the same two basic phenomena: (a) spontaneous generation of wave fronts at the leading centers as described in Section 5 (ii), and (b) their subsequent motion, according to the rules brought out in Section 4. The generation process in 5 (ii) was for pairs of diverging fronts moving in one space dimension; its two-dimensional analog is the spontaneous appearance of a small circular front which spreads outward. Since the fronts are very narrow in our analysis, they appear locally as plane waves. Therefore it suffices to treat the problem in a one-dimensional framework, which we shall do. Alternatively, the variable x could be interpreted as distance to the origin in a configuration with radial symmetry.

(i) Imposed heterogeneities. We suppose that near the origin there is a substance with prescribed density distribution $w(x)$; alternately, w could represent an imposed temperature distribution. We also suppose that w influences the reaction process, so that f and g in (3) are functions of u, v, and w. For each value of w, the nullcurves $f = 0$ and $g = 0$ have the shape shown in Figure 1 ; but their relative positions may vary with w. In particular, $h_{\pm}$, $\bar{v}$, and $\underline{v}$ may depend on w.

At $x = 0$, fronts involving an abrupt increase in u (upjump fronts) form, according to the description in Section 5, when v has a local minimum at the origin which decreases to $\underline{v}$. Similarly, downjump fronts form when $u = h_+(v)$ and v has a maximum which increases to $\bar{v}$. The result is that when x is fixed at 0, the trajectory $(u(0,t), v(0,t))$ is that of a relaxation oscillator with the kinetics of Figure 1. The nullcurve $g = 0$ must be placed as shown, to ensure that the kinetics are oscillatory.

The pattern must be periodic in time. The period T is set by the period of the relaxation oscillatory motion followed by the solution at $x = 0$, $w = w(0)$. This is presumed known.

The mathematical analysis of the above conceptual model consists in determining the trajectories of the expanding fronts, and the function $v(x,t)$, so that all the above evolutionary laws and constraints are fulfilled. It is convenient to express the fronts' motion by the functions $\tau^{\pm}(x)$. Here $\tau^{+}(x)$ is the time at which some specific upjump front reaches position x, and $\tau^{-}(x)_{+}$ is the time for the next succeeding downjump. The next upjump is then at time $\tau + T$. The equations to be satisfied are:

$$\frac{d}{dx}\,\tau^{\pm}(x) = \pm\, c^{-1}(v(x,\tau^{\pm}(x)),w(x)), \quad \text{(for trigger fronts)},$$

$$\frac{d}{dx}\,\tau^{\pm}(x) \quad \text{given by (9) (for phase fronts)},$$

$$\frac{\partial v}{\partial t} = \begin{cases} G_{+}(v,w(x)), \tau^{+}(x) < t < \tau^{-}(x)\ , \\ G_{-}(v,w(x)),\ \bar{\tau}\,(x) < t < \overset{+}{\tau}\,(x) + T \end{cases}$$

(it suffices to determine v in the interval $\tau^{+} < t < \tau^{+} + T$). The periodicity constraint on v is that $v(x,\tau^{+}(v)) = v(x,\tau^{+}(x) + T)$. In addition, we must require that $v(0,t)$ be the values of v corresponding to the relaxation oscillator at the center, and that as $x \to \infty$, the $\tau^{\pm}(x)$ approach linear functions (corresponding to a plane wave train).

If we insist that all the fronts be trigger, then the inequality

$$\underline{v}(w(x)) < v < \bar{v}(w(x))$$

is an additional constraint, and it is a nontrivial matter to determine whether there exist functions v, satisfying all the above. However, the problem becomes somewhat easier when one allows every other front (say, all the down jump fronts) to be of phase type, the remaining trigger. This appears to be the situation commonly observed in the [illegible] targets.

Tyson [1] has produced a scaled version of a set of reaction-diffusion equations (the "Oregonator") realistically modeling the Belousov-Žabotinskii reaction. We [27] have investigated targets with alternating phase and trigger fronts using this system.

The third conceptual possibility is when all fronts are of phase type. Then the configuration of the fronts, as they develop in the reagent, will depend completely on the initial concentrations, and will not in general be circles. Such patterns, if they do exist, would therefore not account for the observed circular fronts.

(ii) Self-sustaining target patterns may be modelled by retaining the function $w(x)$, but supposing it to obey a third reaction-diffusion equation coupled to the first two. Thus the distribution of w is obtained as part of the model, not imposed by external conditions. This type of model is explored in some detail in [17], assuming all fronts are trigger. Again, the dynamics of the wave front and the functions v and w involve complicated mathematics, but some reasonable simplifications are possible.

A conceptually similar model for self-sustaining patterns involving three reacting components had previously been proposed by Zaikin and Kawczyński [28]; but its analysis was left incomplete and the question of phase and trigger fronts was not addressed.

7. Targets in excitable media.

With nullcurves as depicted in Figure 1, the kinetic equations

$$u_t = kf(u,v), \; v_t = g(u,v)$$

have stable relaxation oscillatory solutions. When the g nullcurve is shifted to one of the positions in Figures 3 and 4, however, the kinetics become excitable or bistable.

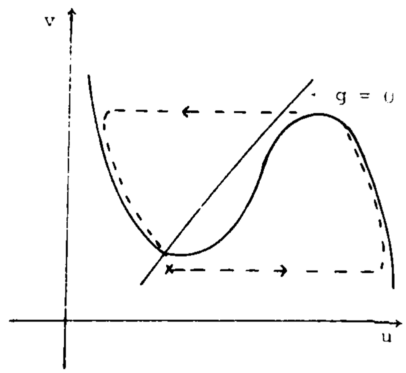

Figure 3

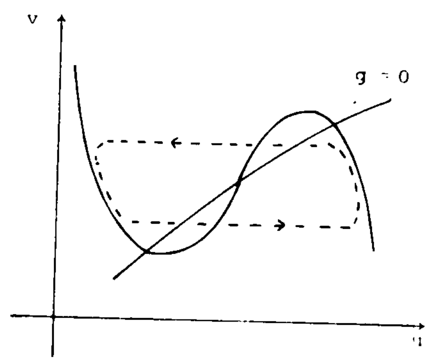

Figure 4

For example in Figure 3, there is one stable rest state (the unique intersection point); but when this state is perturbed downward a small amount, the solution makes a large excursion (dotted line) before returning to the rest state.

In modeling target patterns in Section 6, we needed the kinetics to be oscillatory at the origin; but away from the origin, the configurations in Figures 3 and 4 are not excluded. As $x \to \infty$ the target develops into a regular wave train, so of course the kinetics must support such a train there. Excitable and bistable (as well as oscillatory) kinetics do support wave trains. For example, the phase plane image (orbit) of a train at a fixed value of x is shown by the dotted loop in Figure 4. Therefore there is no contradiction involved in having a periodic target pattern emerge in an excitable medium.

This is apparently what often happens with the Z-reagent, which can exist in an excitable, as well as oscillatory, regime. The initial formation of a target pattern in such a medium involves a wave train entering a quiescant region at a stable rest state. It may be difficult to visualize this process of excitation into a periodic state, but several mechanisms are available for accomplishing it [24; R. Smock, in preparation].

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SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

REPORT DOCUMENTATION PAGE

READ INSTRUCTIONS BEFORE COMPLETING FORM

1. REPORT NUMBER 2074	2. GOVT ACCESSION NO. AD-A089 590
3. RECIPIENT'S CATALOG NUMBER	
4. TITLE (and Subtitle) PROPAGATING WAVES AND TARGET PATTERNS IN CHEMICAL SYSTENS	5. TYPE OF REPORT & PERIOD COVERED Summary Report - no specific reporting period
	6. PERFORMING ORG. REPORT NUMBER
7. AUTHOR(s) Paul C. Fife and John Tyson	8. CONTRACT OR GRANT NUMBER(s) DAAG29-80-C-0041, DAAG29-75-C-0024, MCS79-04443
9. PERFORMING ORGANIZATION NAME AND ADDRESS Mathematics Research Center, University of Wisconsin, 610 Walnut Street, Madison, Wisconsin 53706	10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS 2 - Physical Mathematics
11. CONTROLLING OFFICE NAME AND ADDRESS See 18 below	12. REPORT DATE May 1980
	13. NUMBER OF PAGES 19
14. MONITORING AGENCY NAME & ADDRESS(if different from Controlling Office)	15. SECURITY CLASS. (of this report) UNCLASSIFIED
	15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)

Approved for public release; distribution unlimited.

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

18. SUPPLEMENTARY NOTES

U. S. Army Research Office
P.O. Box 12211
Research Triangle Park,
North Carolina 27709

National Science Foundation
Washington, D.C. 20550

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

Reaction, Diffusion, Chemical waves, Wave front, Pattern formation, Singular perturbations

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

A class of models for target patterns (concentric circular waves emanating from a point called the leading center) is constructed in the context of singularly perturbed reaction-diffusion systems of partial differential equations. First, the theory of wave fronts is detailed for scalar equations and systems of equations. A scaling method reduces complex waves to the consideration of a group of simple wave phenomena. It is shown that expanding wave fronts can be generated spontaneously at a point. This process, together with the laws of their subsequent motion, reduces the problem to an ordinary differential initial

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20 Abstract (continued)

value problem, whose solution is required to have certain properties. A discussion is given of the connection between these results and experimental observations.