Turbulent Spatio-Temporal Dynamics in Reacting-Diffusing Systems: Results for a Soluble Model

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A class of soluble three "species" reaction-diffusion type systems is presented Exact solutions are obtained which show turbulent spatio-temporal evolution All homogeneous evolution tends asymptotically toward an attractor which is shown to be a two layered two dimensional manifold in the three dimensional species space. Sustained aperiodic spatio-temporal solutions are also found.

By considering particular model systems we show that turbulent solutions may exit as finite amplitude instabilities or as bifurcations which are aperiodic arbitrarily close to the bifurcation point and hence do not arise as a transition starting out essentially periodically.

A perturbation scheme is used to show that d parameter families of spatio-temporal evolution are admitted by more general systems with attracting d dimensional manifolds in the homogeneous chemical kinetics.

I. Introduction

Although sustained aperiodic motion has been long discussed in hydrodynamic systems in the study of turbulence [1, 2], such evolution has only recently been found in chemically reacting systems. Reaction models involving coupled oscillatory systems leading to quasiperiodic motion [3], and a variety of reaction kinetic topologies leading to chaos have been pointed out [4]. Using perturbation techniques to study the inhomogeneous evolution of systems with a homogeneous limit cycle oscillation, it was pointed out that multiply periodic inhomogeneous evolution could arise in reacting diffusing systems [5].

All of the above developments [3, 4] have been carried out either by numerical simulations or, as in Ref. [5], by an asymptotic expansion technique. Here we consider a simple set of model dynamical equation which can be solved and whose solutions demonstrate sustained homogeneous aperiodic temporal evolution on an attracting surface in concentration space whose analytical form can be specified. Exact solutions of spatio-temporal aperiodicity are found and shown to represent two parameter families of such solutions.

In the last section we investigate the effect of adding diffusion to a system having a d-dimensional attractor in the chemical kinetics. A formal perturbation theory is used to show that a multiparameter family occurs as an extension of the homogeneous evolution to weakly spatially varying aperiodic solutions for certain classes of "well behaved" attractors. The theory appears to break down for "strange" attractors and some conjectures on the implications of this break-down are made.

II. The "Polarator" Model

In the work of several authors on limit cycles and wave propagation [6–8] a class of soluble two species dynamical models was studied. The feature which made the models tractable was the fact that when the descriptive variables X, Y were transformed to two dimensional polar variables ($X = R\cos\theta$, $Y = R\sin\theta$) the angular and radial coordinates obeyed a particularly simple set of kinetic equations ($\mathrm{d}R/\mathrm{d}t = R\,B(R)$, $\mathrm{d}\theta/\mathrm{d}t = A(R)$) for homogeneous evolution. In this section we consider an extension of this concept to three dimensional polar variables R, θ, Φ as defined in Figure 1. The polar variables are taken to obey the following set

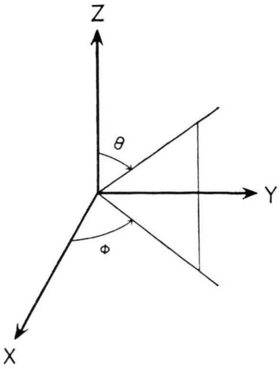

Fig. 1. Coordinate systems for the polarator.

of dynamical equations defining our system,

$$\mathrm{d}R/\mathrm{d}t = R\,B(R)\,, \tag{II.1}$$

$$\mathrm{d}\theta/\mathrm{d}t = T(R)\,, \tag{II.2}$$

$$\mathrm{d}\varphi/\mathrm{d}t = P(R)\,. \tag{II.3}$$

Using the polar transformation

$$\begin{aligned} X &= R \sin\theta \cos\varphi\,, \\ Y &= R \sin\theta \sin\varphi\,, \\ Z &= R \cos\theta\,, \end{aligned} \tag{II.4}$$

we obtain a set of model reaction diffusion equations

$$\frac{\partial X}{\partial t} = D_X \nabla^2 X + BX + \frac{XZT}{(X^2+Y^2)^{1/2}} - PY\,, \tag{II.5}$$

$$\frac{\partial Y}{\partial t} = D_Y \nabla^2 Y + BY + \frac{YZT}{(X^2+Y^2)^{1/2}} + PX\,, \tag{II.6}$$

$$\frac{\partial Z}{\partial t} = D_Z \nabla^2 Z + BZ - (X^2+Y^2)^{1/2}\,T \tag{II.7}$$

for the dynamics in terms of the cartesian variables X, Y, Z with diffusion coefficients D_X, D_Y and D_Z respectively.

Homogeneous Evolution

Clearly if

$$\lim_{R\to 0} R\,B(R) = 0\,, \tag{II.8}$$

the system has a steady state at the origin, $X = Y = Z = 0$. For cases wherein the property

$$\mathrm{d}/\mathrm{d}R(R\,B(R)) > 0 \quad \text{at} \quad R = 0 \tag{II.9}$$

the steady state at the origin is unstable. We assume there is at least one nonzero value of R, denoted R_0, such that

$$B(R_0) = 0\,, \tag{II.10}$$

$$\mathrm{d}B(R)/\mathrm{d}R < 0 \quad \text{at} \quad R = R_0\,. \tag{II.11}$$

The condition (II.10) allows for the existance of a steady state of the R Eq. (II.1) while (II.11) insures its stability. Thus for the polarator there is a domain $R_n < R < R_x$ such that

$$\begin{aligned} R\,B(R) &> 0\,, \quad R_n < R < R_0\,, \\ R\,B(R) &< 0\,, \quad R_0 < R < R_x\,. \end{aligned} \tag{II.12}$$

For example the simple case $B = 1 - R^2$ obey all the above requirements on B and, in particular, $R_0 = 1$. Other specific examples demonstrating a variety of phenomena will be considered later.

With these assumptions it is clear that there exists a class of solutions such that if

$$R_n < R(0) < R_x\,, \tag{II.13}$$

i.e. if the initial value is in the domain of attraction of the sphere $R = R_0$, then

$$\left.\begin{aligned} R(t) &\sim R_0\,, \\ \theta(t) &\sim \alpha + T_0 t \\ \varphi(t) &\sim \beta + P_0 t \end{aligned}\right\} \quad \text{as} \quad t \to \infty$$

where the frequencies T_0 and P_0 are $T(R_0)$ and $P(R_0)$ respectively and α and β are constant phases. These asymptotic solutions rotate around the invarient sphere with frequencies of rotation T_0 and P_0 with respect to the θ and φ variations respectively. A typical example of such a trajectory is shown in Figure 2. Note that only for the very special case $nT_0 = mP_0$ (where n and m are integers) will the motion be periodic. Otherwise the noncommensurate motion of θ and φ leads to aperiodic motion which eventually fills the sphere (as we shall show below).

The question arises as to how motion on a two dimensional manifold, the sphere $R = R_0$, can lead to intersecting trajectories and we now turn to this point.

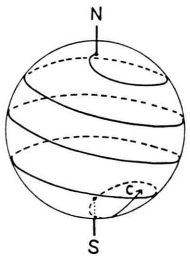

Fig. 2. Typical asymptotic trajectory for the polarator on the invarient sphere showing nonuniqueness at points such as C where the trajectory crosses itself.

III. Nonuniqueness of the Orbits of the Polarator Attractor

A) Distortion of the Invarient Torus

We now investigate the qualitative aspects of an alternative model which we then map into the polarator system by deformation of its attractor.

Consider the torus shown in Fig. 3 and assume we construct a system with it as attractor. A typical asymptotic trajectory is shown and we assume that, like the polarator, the winding around the torus is at a noncommensurate frequency to that of the rotation around the vertical axis.

In Fig. 4 we show a sequence of deformations of the torus into a two layered spherical shell with holes at the north and south poles. Clearly the toroidal model can generate the polarator dynamics to any degree of accuracy as we deform it to have vanishing thickness and holes at the north and south poles of vanishing area.

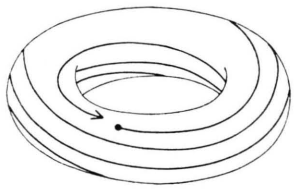

Fig. 3. Toroidal attractor with an aperiodic orbit showing uniqueness of the trajectories which always manage to avoid each other by winding around the torus in the same sense.

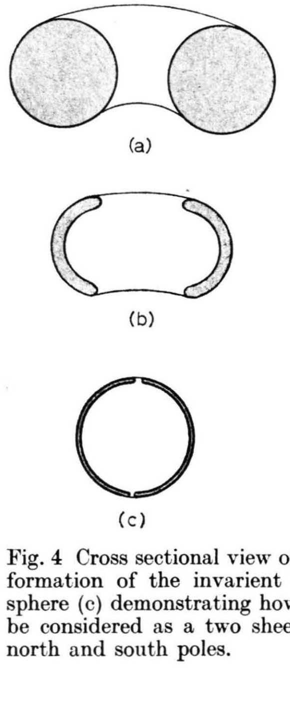

Fig. 4 Cross sectional view of the progressive (a) $\rightarrow$ (c) deformation of the invarient torus (a) into the invarient sphere (c) demonstrating how the polarator attractor may be considered as a two sheeted sphere with holes at the north and south poles.

The importance of this distortion, torus $\Rightarrow$ polarator, is that the deformed torus is a two dimensional attractor with unique trajectories while the limiting case, the polarator, has nonunique (intersecting) trajectories.

The reason why the polarator gives nonunique orbits (e.g. the possibility of orbits crossing as at point C in Fig. 2) is that there is a multiple valuedness in the polar transformation. The coordinate θ is usually defined to be between 0 and π (and similarly $0 < \varphi < 2\pi$). However as θ increases beyond π it certainly generates unique orbits in the R, θ, φ variables but must be considered to transform to a new sphere — for example, the inside layer of the distorted torus — when understanding the trajectories in the X, Y, Z variables.

B) Breakdown of the Criteria for Uniqueness

Let us now take another point of view on the question of uniqueness, showing that the usual condition guaranteeing this property fails for the dynamics of the homogeneous evolution in the cartesian variables. The usual sufficient conditions for existence and uniqueness of the solutions of a set of ordinary differential equations requires that the rate law (F) satisfy a Lipschitz condition [12]. For the set of Eqs. (III.5—7) this is not satisfied since the limit of the terms containing $(X^2 + Y^2)^{1/2}$ in (III.5, 6) as we approach the poles depends on the direction of approach (e.g. the values of X/Y as $X, Y \rightarrow 0$; the rate law is not continuous.

IV. Space Filling Property of the Polarator Orbits on the Sphere

It is easy to show for spatially homogeneous motion that any orbit on the sphere can become arbitrarily close to any specified point on the sphere. Consider a point θ_0, φ_0 on the sphere,

$$0 \leqq \theta_0 \leqq \pi\,, \quad 0 \leqq \varphi < 2\pi\,. \tag{IV.1}$$

To demonstrate that the asymptotic trajectories fill the sphere we simply must show that for sufficiently large t

$$\left| \underset{\pi}{\text{mod}} \{\alpha + T_0 t\} - \theta_0 \right| < \varepsilon\,, \tag{IV.2}$$

$$\left| \underset{2\pi}{\text{mod}} \{\beta + P_0 t\} - \varphi_0 \right| < \varepsilon \tag{IV.3}$$

for arbitrarily small ε. The notation $\underset{Q}{\mathrm{mod}}$ means the value of the angle modulo a multiple of the angle Q.

Clearly one of the conditions can always be met. Defining t_n such that

$$\underset{\pi}{\mathrm{mod}}[\alpha + T_0 t_n] = \theta_0 \tag{IV.4}$$

the condition (IV.2) is satisfied for $\varepsilon = 0$. Letting

$$t_n = (n\pi + \gamma)/T_0, \quad n = 0, \quad \pm 1, \ldots, \tag{IV.5}$$

we see that the angle γ is fixed by the equations

$$\underset{\pi}{\mathrm{mod}}[\alpha + \gamma] = \theta_0, \quad 0 < \gamma < \pi. \tag{IV.6}$$

Now the problem reduces to showing that we can always find an n so that (IV.3) is satisfied for arbitrarily small ε.

Let I_n be defined as

$$I_n = \underset{2\pi}{\mathrm{mod}}\{\beta + P_0 t_n\}. \tag{IV.7}$$

Thus

$$I_n = \underset{2\pi}{\mathrm{mod}}\{\beta' + n P_0 \pi/T_0\} \tag{IV.8}$$

where β' depends on β and on α (through γ),

$$\beta' \equiv \beta + P_0 \gamma/T_0. \tag{IV.9}$$

For the case where P_0 and T_0 cannot be written as a ratio of integers, the noncommensurate case, the number $\underset{2\pi}{\mathrm{mod}}\{n P_0 \pi/T_0\}$ can get arbitrarily close to any number in the internal $[0, 2\pi]$ for sufficiently large n and hence (IV.2, 3) can be satisfied for arbitrarily small ε if we wait sufficiently long. Thus the polarator trajectories for homogeneous evolution fill the invarient attractor $R = R_0$.

V. Two Parameter Families of Spatio-Temporarily Aperiodic Solutions

Transforming the reaction-diffusion Eqs. (II.5—7) to the polar variables R, θ, φ we have been able to obtain a two parameter family of spatio-temporarily aperiodic solutions for the case of equal diagonal diffusion,

$$D_i = D, \quad i = X, Y, Z \tag{V.1}$$

in an infinite one dimensional medium or system with periodic boundary conditions (i.e. a ring) along a spatial variable r. We find solutions of the form

$$R = R_k, \tag{V.2}$$

$$\theta = \bar{\theta} + T(R_k)t + kr, \tag{V.3}$$

$$\varphi = \bar{\varphi} + P(R_k)t, \tag{V.4}$$

where $\bar{\theta}$ and $\bar{\varphi}$ are arbitrary constants and R_k is the solution of

$$B(R_k) - k^2 D = 0 \tag{V.5}$$

as may be varified by direct substitution.

Using the elementary identity $2 \sin A \cos B = \sin(A + B) + \sin(A - B)$ the cartesian variable X thus evolve according to

$$X(r,t) = \tfrac{1}{2} R_k \sin[\alpha_+ + w_+(k)t + kr] + \tfrac{1}{2} R_k \sin[\alpha_- + w_-(k)t + kr] \tag{V.6}$$

where

$$\alpha_\pm = \bar{\theta} \pm \bar{\varphi}, \tag{V.7}$$

$$w_\pm(k) = T(R_k) \pm P(R_k) \tag{V.8}$$

and similarly for Y and Z.

A) Two Parameter Family of Solutions

From (V.6) it is clear that the solutions presented are pairs of waves with frequencies $w_\pm(k)$ of wavevector k. Since the system is infinite, translational invariance demands that we can always construct a new solution by a shift of coordinates $r \to r + r_0$. Fixing this coordinate system so that $kr_0 + \alpha_+ = 0$ we obtain

$$X(r,t) = \tfrac{1}{2} R_k \sin[w_+(k)t + kr] + \tfrac{1}{2} R_k \sin[w_-(k)t + kr + \alpha], \tag{V.9}$$

where $\alpha = \alpha_- - \alpha_+$. Clearly even after the translational invariance degree of freedom has been eliminated there are still two parameters fixing the family of solutions:

(1) the wavevector k,

(2) the phase angle α between the $+$ and $-$ waves.

B) Space Filling Dynamics

By an identical argument as that presented in Section IV one may show that each member (α) of the two parameter family for a given k fills the sphere of radius R_k (except, of course, at those isolated values of k for which $w_+(k)/w_-(k)$ is a rational fraction).

C) Stability of the Family

The members of the two parameter family are at most marginally stable as follows: Clearly they are marginally stable to a small overall translation of the wave $r \to r + \delta r$ because of translational invariance of the infinite medium (or ring shaped vessel). However it is obvious that if $X(r, t\,|\,\alpha, k)$ in (V.9) is a solution for the system then $(\partial X(r, t\,|\,\alpha k)/\partial\alpha)$ is a solution of the equations linearized about $X(r, t\,|\,\alpha, k)$ and hence since $(\partial X/\partial\alpha)$ does not decay in time X is only marginally stable to a perturbation, e.g. $\delta(X, Y, Z) \propto \partial/\partial\alpha[X, Y, Z]$. This marginal stability is due to the fact that every solution is embedded in a continuum of solutions (parametrized by α). Indeed even the homogeneous asymptotic evolution on the invarient sphere is parametrized by a $\theta - \varphi$ phase shift (in addition to a time translation along any trajectory) giving rise to a one parameter family of homogeneous aperiodic colutions.

It seems intuitively reasonable to suggest that the aperiodic solutions studied here will be stable in the sense that a small perturbation from one of them will at most lead asymptotically ($t \to \infty$) to another member of the family. This has in fact been shown for one dimensional attractor', in the case of the stable limit cycle [9].

D) Bifurcation Directly into Aperiodic Behavior

Let us consider the specific example [5],

$$B = 1 - R^2. \tag{V.10}$$

For this system the spatio-temporal evolution at wave vector k takes place on a sphere of radius R_k given by

$$R_k = \sqrt{1 - k^2 D}. \tag{V.11}$$

The system has solutions of the type studied above for

$$0 \leqq |k| < k_c \equiv D^{-1/2}. \tag{V.12}$$

Clearly the family terminates in a bifurcation at $|k| = k_c$ with bifurcation index 1/2. It is interesting to note that the temporal evolution of even the vanishingly small amplitude solutions, $k \lesssim k_c$, are still aperiodic and thus the bifurcation does not start out periodically.

E) Aperiodic Evolution as a Finite Amplitude Instability

Taking the case [5]

$$B = 1 - (\varrho^2 - R^2)^2 \tag{V.13}$$

we obtain two solutions (taking $D_i = 1$ for simplicity)

$$R_{k,\pm} = [\varrho^2 \pm (1 - k^2)^{1/2}]^{1/2} \tag{V.14}$$

as shown in Figure 5. Note that for the case $\varrho > 1$ the system has a stable homogeneous state $R = 0$ but may be excited to the various aperiodic solutions at R_k by a finite amplitude perturbation.

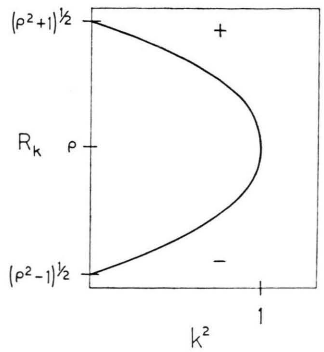

Fig. 5. Bifurcation diagram for the case of a finite amplitude transition to temporal chaos as discussed in Section VE.

F) Other Phenomena

One may construct a variety of examples to demonstrate various types of dispersion relations $w_\pm(k)$ and families of waves with other analytical behavior in the amplitude R_k. The results are essentially the same as given in Ref. [5] where the soluble X, $Y \to R$, θ model was discussed.

G) More General Solutions for the Case T, P Constant, $D_X = D_Y \neq D_Z$

For cases where the frequencies T and P are independent of R we have formed solutions for the case $D_X = D_Y \neq D_Z$. Consider solutions of the form

$$\begin{aligned} X &= R(t)\sin\theta\cos\varphi\,, \\ Y &= R(t)\sin\theta\sin\varphi\,, \\ Z &= R(t)\cos\theta\,, \end{aligned} \tag{V.15}$$

where

$$\begin{aligned} \theta &= \bar\theta + Tt + \boldsymbol{k}\cdot\boldsymbol{r}\,, \\ \varphi &= \bar\varphi + Pt + \boldsymbol{q}\cdot\boldsymbol{r}\,, \\ \boldsymbol{k}\cdot\boldsymbol{q} &= 0\,. \end{aligned} \tag{V.16}$$

Again $\bar\theta$ and $\bar\varphi$ are constants. The wave vectors $\boldsymbol{k}$ and $\boldsymbol{q}$ are orthogonal. It is easy to show that these solutions satisfy the reaction diffusion Eqs. (II.5–7) when

$$\mathrm{d}R/\mathrm{d}t = (B(R) - k^2 D_Z)\,R\,, \tag{V.17}$$

$$q^2 = k^2(D_Z/D_X - 1)\,. \tag{V.18}$$

These solutions only arise when

$$D_Z \geqq D_X \tag{V.19}$$

since otherwise $q^2 < 0$ and the solutions (V.15, 16) are unbounded at infinity.

From (V.17) it is clear that constant amplitude (R) solutions exist at values R_k of R such that

$$B(R_k) - k^2 D_Z = 0\,. \tag{V.20}$$

These steady amplitude solutions are attracting with respect to the general class of solutions of the type (V.15, 16) if

$$(\mathrm{d}B/\mathrm{d}R)_{R=R_k} < 0\,. \tag{V.21}$$

It must be stressed that this condition is not equivalent to saying that the solutions (of the type (V.15, 16) with R taking the value R_k given by (V.20)) are attractors of the full system since we have not shown that small perturbations from these solutions which are not of the form (V.15, 16) also approach the R_k states as $\mathrm{t} \to \infty$.

Using (V.21) one may verify that for the case $B = 1 - R^2$ the solutions at $R_k = (1 - k^2 D_Z)^{1/2}$ are attracting in the above sense for all values of k for which the solutions exist ($0 \leqq k^2 \leqq D_Z{}^{-1}$). Similarly for the case $B = 1 - (\varrho^2 - R^2)^2$ the solutions with $R_{k,+}$ given by (V.14) are attracting (in the above sense) while the solutions with $R_{k,-}$ are unstable.

H) Standing Waves for the Case T, P Independent of R, $D_i = D$

One may find a class of solutions to the reaction diffusion system in the form

$$\begin{aligned} X &= R(\boldsymbol{r}) \sin(\bar{\theta} + T t) \cos(\bar{\varphi} + P t)\,,\\ Y &= R(\boldsymbol{r}) \sin(\bar{\theta} + T t) \sin(\bar{\varphi} + P t)\,,\\ Z &= R(\boldsymbol{r}) \cos(\bar{\theta} + T t)\,. \end{aligned} \tag{V.22}$$

With this one finds that $R(\boldsymbol{r})$ must obey the equation

$$D \nabla^2 R + R B(R) = 0\,. \tag{V.23}$$

Consider the case $B = 1 - \gamma R^2$ in one dimension r. We obtain ($\mathrm{d}/\mathrm{d}r = {}'$)

$$D R'' + R \pm \gamma R^3 = 0\,. \tag{V.24}$$

This is the classical equation for a nonlinear spring. When γ is small the solutions to (V.24) are of the form [13]

$$R = a \cos kr + (\gamma a^3/32) \cos 3kr + O(\gamma^2) \tag{V.25}$$

where a is an arbitrary (but small) amplitude and the wave vector k is given by

$$D^{1/2} k(a, \gamma) = \pm [1 - (3a^2/8)\gamma + O(\gamma^2)]\,. \tag{V.26}$$

An alternative interpretation of this latter equation is to determine the dependence of the amplitude a as a function of the wave vector k. From (V.26) we obtain

$$a = \left[\frac{4(1 - k^2 D)}{3\gamma}\right]^{1/2}.$$

Thus these standing waves correspond to a family, parametrized by the wave vector k, which exist in a neighborhood of K values near but less than $D^{-1/2}$ (for $\gamma > 0$).

I. Ring Reactors

Clearly in a ring of circumference L we must have $\Psi(r + L) = \Psi(r)$ and hence $kL = 2n\pi$ where n is an integer. The solutions presented above are valid at wave vectors k_n obeying both the cutoff restrictions and the quantization condition

$$k_n = 2n\pi/L\,, \quad n = 0, \pm 1, \ldots. \tag{V.27}$$

Thus for a system with a cut off such as in Sect. VD we have inhomogeneous solutions for

$$L > 2\pi/k_{\mathrm{c}} \tag{V.28}$$

where k_{c} is the maximum wavevector for which the system may sustain solutions.

VI. Long Length Scale Extensions for More General Attractors

A) Basic Equations and Assumptions

We consider a general kinetic system

$$\partial\Psi/\partial t = \mathscr{D} \nabla^2 \Psi + \mathscr{F}[\Psi] \tag{VI.1}$$

and assume that the associated ordinary differential equation

$$\mathrm{d}\Psi/\mathrm{d}t = \mathscr{F}[\Psi] \tag{VI.2}$$

has an attractor such that all solutions in some nonzero domain evolve to a bounded d dimensional manifold. We further assume that all asymptotic solutions on the manifold may be characterized by d parameters, one of which corresponds to a displacement in time. The question arises as to what is the relationship between the attractors of (VI.2) and the reaction-diffusion system (VI.1). In this section we address this question in the case where diffusion plays a perturbative role, i.e. the solutions are on a long length scale.

Let t_c be a typical time scale embedded in $\mathscr{F}$ and $\mathscr{D}_c$ be a characteristic diffusion coefficient. With this we may construct a characteristic length L_c according to

$$L_c^2 = \mathscr{D}_c t_c \,. \tag{VI.3}$$

If we further introduce the typical length L of our solution we may define dimensionless space ($\boldsymbol{\rho}$) and time (τ) variables,

$$\boldsymbol{\rho} = \boldsymbol{r}/L \,, \quad \tau = t/t_c \,. \tag{VI.4}$$

Letting the laplacian in the scaled variable be denoted ∇^2 for simplicity, (IV.1) takes the scaled form

$$\partial \Psi/\partial \tau = \varepsilon D \nabla^2 \Psi + F(\Psi) \tag{VI.5}$$

with

$$\mathscr{D} = D_c D \,, \quad \mathscr{F} = t_c^{-1} F \,. \tag{VI.6}$$

The length scale parameter ε is given by

$$\varepsilon = (L_c/L)^2 \ll 1 \tag{VI.7}$$

and is taken to be small. Thus the meaning of smooth spatial variation is made more precise, e.g. on a scale much longer than the intrinsic length L_c of the reaction-diffusion phenomenological relations.

This formulation of the problem naturally suggests an asymptotic development in ε as $\varepsilon \to 0$. In earlier work on the extension of limit cycles to plane and other geometrical wave forms [5, 11] it was shown that one must allow for renormalization of the time due to the effects of diffusion. More generally diffusion introduces multiple time scales (slower than t_c) into the problem for spatially smooth solutions. Thus we introduce the times τ_n, such that

$$\tau_n = \varepsilon^n \tau \,, \quad n = 0, 1, 2, \ldots \,. \tag{VI.8}$$

Furthermore the phase of oscillation, a parameter characterizing the solution on the one dimensional limit cycle attractor, could depend on space. The natural extension of this idea is that the parameters ζ characterizing the trajectories on the d dimensional attractor, will be dependent on the spatial coordinate $\boldsymbol{\rho}$ and the slow times τ_n, $n \geqq 1$. We shall now consider a formal development of Ψ and the inhomogeneous time dependent extension of the parameters ζ in a series expansion in ε.

B) The Formal ε Development

We start by assuming the existence of an expansion of Ψ in the form

$$\Psi = \sum_{n=0}^{\infty} \Psi_n \varepsilon^n \,. \tag{VI.9}$$

$$\partial/\partial \tau = \sum_{n=0}^{\infty} \varepsilon^n \, \partial/\partial \tau_n \,. \tag{VI.10}$$

These ε developments are inserted into the scaled Eq. (VI.5).

To lowest order we obtain

$$\partial \Psi_0/\partial \tau_0 = F(\Psi_0) \tag{VI.11}$$

and from our assumption that the solutions be near the attractor $\mathscr{A}$ of F we have

$$\Psi_0 = \varphi(\tau_0, \zeta_0(\boldsymbol{\rho}, \tau_1, \tau_2, \ldots)) \tag{VI.12}$$

where $\varphi(\tau, \zeta)$ is a trajectory on $\mathscr{A}$. From our discussion and assumption of this section φ is parametrized by a set of constants ζ which may, for $\varepsilon > 0$, be expected to depend on space $\boldsymbol{\rho}$ and the slow times $\tau_{n \geqq 1}$. Note that we have implicitly assumed that the spatio-temporal extension of the parameter ζ may be written as a development in ε similar to (VI.9). We now proceed to show that ζ_0 is determined in the next order.

Collecting terms proportional to ε we have

$$\frac{\partial \Psi_0}{\partial \tau_1} + \frac{\partial \Psi_1}{\partial \tau_0} = D \nabla^2 \Psi_0 + \Omega(\Psi_0) \Psi_1 \tag{VI.13}$$

where the matrix Ω is defined by

$$\Omega(\Psi) = \partial F/\partial \Psi \,. \tag{VI.14}$$

Since Ψ_0 depends on $\boldsymbol{\rho}$ through ζ_0 we have

$$\nabla^2 \Psi_0 = \sum_{i=1}^{d} \frac{\partial \Psi_0}{\partial \zeta_{0i}} \nabla^2 \zeta_{0i} + \sum_{i,j=1}^{d} \frac{\partial^2 \Psi_0}{\partial \zeta_{0i} \, \partial \zeta_{0j}} \cdot \boldsymbol{\nabla} \zeta_{0i} \cdot \nabla \zeta_{0j} \,. \tag{VI.15}$$

To proceed further we must determine under what conditions (VI.13) has a solution for Ψ_1. These conditions, indeed, yield equations for the ζ_{0i}.

First we note that by taking the derivative of (VI.11) with respect to ζ_i we have

$$\mathscr{L}(\partial \Psi_0/\partial \zeta_i) = 0 \tag{VI.16}$$

where the linear operator $\mathscr{L}$ is defined by

$$\mathscr{L} \equiv (\partial/\partial \tau) - \Omega(\Psi_0) \,. \tag{VI.17}$$

Thus it is clear that $\mathscr{L}$ has a d dimensional null space spanned by the functions $(\partial \Psi_0/\partial \zeta_{0i})$, $i = 1$,

$2, \ldots, d$. Rewriting (VI.14) in the form

$$\mathscr{L}\Psi_1 = \sigma, \tag{VI.18}$$

$$\sigma \equiv D\nabla^2\Psi_0 - \partial\Psi_0/\partial\tau_1 \tag{VI.19}$$

it is seen that in order that we may determine Ψ_1, the source term σ defined in (VI.19), must not have a projection into the null space of $\mathscr{L}$. This orthogonality yields d equations with which we can determine the d functions ζ_{0i}.

For simplicity we assume that we may introduce an inner product in the space in which $\mathscr{L}$ operates. Taking $\{u_1, \ldots, u_d\}$ to be a complete set of eigenvectors (of the adjoint of $\mathscr{L}$) with zero eigenvalue, the conditions for solubility for Ψ_1, e.g. that σ has no projection in the null space of $\mathscr{L}$, becomes

$$\frac{\partial\zeta_{0i}}{\partial\tau_1} = \sum_{j=1}^{d} A_{ij}\nabla^2\zeta_{0j} + \sum_{j,k=1}^{d} B_{ijk}\nabla\zeta_{0j}\cdot\nabla\zeta_{0k}$$

where

$$A_{ij}(\zeta_0) \equiv \left(u_i, D\frac{\partial\varphi}{\partial\zeta_{0j}}\right), \tag{VI.21}$$

$$B_{ijk}(\zeta_0) \equiv \left(u_i, D\frac{\partial^2\varphi}{\partial\zeta_{0j}\,\partial\xi_{0k}}\right) = B_{ikj}(\zeta_0), \tag{VI.22}$$

$$\left(u_i, \frac{\partial\varphi_0}{\partial\zeta_{0j}}\right) = \delta_{ij}. \tag{VI.23}$$

We have assumed biorthonormality via (VI.23). The solubility conditions provide d equations for the d quantities $\zeta_{0i}\,(i = 1, 2, \ldots, d)$.

For the limit cycle, $d = 1$, there is only one ζ parameter corresponding to the phase of oscillation. For that case (VI.21) reduces to an equation for the local phase of oscillation $\zeta_{01}(\varrho, \tau_1, \tau_2, \ldots)$ and has been studied in Ref. [5] and [11].

By their definition the variables ζ_i for homogeneous evolution parametrize the orbits $\varphi(\tau, \zeta)$ on the attractor $\mathscr{A}$. Hence they must be within certain allowed ranges. For example the variables which are phase like in nature (in analogy to θ and φ in the polarator) then they may take on all real values. However certain parameters may be limited to finite real domains and hence allowable solutions must comply to this restriction.

We note the complex structure of the spatio-temporal pattern that arise because of the solutions to (VI.20). As in the polarator (and in contrast to the limit cycle) the phenomena do not typically correspond to an inhomogeneous distribution in the phase of the homogeneous evolution. For the polarator this would correspond to putting $t \to t + kr$ in (V.4) as well as in (IV.3). For $d \geqq 2$, the inhomogeneous evolution may lead to spatio-temporally varying extensions of more than one ζ_i, the parameters of the orbits on the attractor (as for the polarator in the case studied in Section V G).

An important question in understanding these systems is to characterize the asymptotic, $t \to \infty$, properties of the solutions. Consider the solutions of (VI.21) as $\tau_1 \to \infty$. For the polarator we have

$$\zeta_\theta \underset{k\to 0}{\sim} \bar{\theta} + kr + T'(R_0)\left(\frac{\mathrm{d}R_k}{\mathrm{d}k^2}\right)_{k^2=0} k^2 t$$

$$\zeta_\phi \underset{k\to 0}{\sim} \varphi + P'(0)\left(\frac{\mathrm{d}R_k}{\mathrm{d}k^2}\right)_{k=0} k^2 t.$$

Note the system has three parameters, $\bar{\theta}$, $\bar{\varphi}$ and k^2, characterizing the solution. More generally we expect that there will be $d+1$ parameters characterizing the asymptotic spatio-temporal solutions extended from the d dimensional attractor. Surface jumping may bring in other length scales and break down of the theory would occur.

VII. Conclusions

We have shown by the use of a soluble model, the polarator, and by the general plausibility argument of Section VI that one may expect multi-parameter families of spatio-temporal solutions to reaction diffusion systems whose reaction kinetics admits a multiple dimensional attracting manifold. Indeed we conjecture that for a wide class of systems with a d dimensional attractor there will be d parameter families of such solutions (actually $d+1$ if we include a simple shift of the time axis). In future developments we shall present more precise criteria for the existenc and stability of these solutions in general classes of systems with attractors in their chemical kinetics and furthermore apply the theory to the recently found chaotic behavior in the Belousov-Zhabotinsky-Zaikin reaction [10].

We note that the theory developed in Section VI is limited to systems with essentially one time and length scale. Indeed it has been shown using catastrophe theory and matched asymptotic expansions [14, 15] that when multiple time or length scales are of importance even simple attractors such as steady states can lead to a great variety of propagating phenomena. In the presence of multidimensional attractors the range of possibilities is greatly increased and should lead to the discovery of a host of interesting phenomena in the near future.

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