A New Perturbation Approach to Highly Nonlinear Chemical Oscillation with Diffusion Process

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November 4, 1975

We present in this short note a simple perturbation scheme to derive a reduced description of the dynamics of reaction-diffusion systems deep in a temporally ordered state. This work was motivated by Ortoleva and Ross' phase-wave theory of heterogeneous reaction-diffusion system in which the degree of heterogeneity is taken as a small parameter. In contrast to their work, however, we consider a homogeneous system and the amplitude and the instantaneous frequency are expanded in powers of space-derivatives.

We restrict our consideration to a two-component reaction-diffusion system because in this case one may give an explicit definition of phase and amplitude so that the theory may be developed in a most unambiguous way.

We start with the equations

\[ \dot{x} = D_x p \Delta x + F_x(x, y), \]

\[ \dot{y} = D_y p \Delta y + F_y(x, y), \]

(1)

where \( x \) and \( y \) are concentrations of reactants subtracted by steady state values. We are now concerned with the oscillation of \( x \) and \( y \) around \( x = y = 0 \). In such a situation it is natural to introduce a complex variable \( w = w_1 + iw_2 = \rho \exp(i\theta) \), where \( (w_1, w_2) \) is defined by an arbitrary linear transformation of \( (x, y) \). An essential point in our theory is that the nonlinear oscillation of concentrations with slow spatial variation may be described by the equation of motion for phase \( \theta \) in a closed form.

Let us begin with the uniform case and rewrite (1) in the forms

\[ \dot{\rho} = F_\rho(\rho, \theta), \]

\[ \dot{\theta} = F_\theta(\rho, \theta). \]

(2)

By eliminating \( dt \) from (2) a family of trajectories

\[ \rho = \rho_0(\theta) \]

(3)

may in principle be determined. As another independent equation we adopt the second equation in (2). Then, on using (3), one obtains

\[ \dot{\theta} = F_\theta(\rho_0(\theta), \theta) \equiv \Omega_\theta(\theta), \]

(4)

which has a closed form in \( \theta \). It is essential to notice that an integration constant included in \( \rho_0(\theta) \) and hence in \( \Omega_\theta(\theta) \) becomes unimportant in the temporally ordered state. This is due to the fact that the trajectories with various initial conditions will rapidly converge into the same closed orbit. Therefore, except for some initial period, \( \rho_0(\theta) \) and \( \Omega_\theta(\theta) \) may practically be regarded as definite 2\( \pi \)-periodic functions of \( \theta \). At the same time one has

\[ x = x_0(\theta), \quad y = y_0(\theta), \]

(5)

where \( x_0 \) and \( y_0 \) may as well be regarded as 2\( \pi \)-periodic in \( \theta \). Equations (5) and (4) constitute a natural representation of the nonlinear oscillation which is uniform in space.

The above scheme may be generalized to nonuniform case. Assuming that the spatial variation in \( \theta \) is slow and replacing \( p \) by \( e^p \), we shall find \( \rho \) and \( \dot{\theta} \) in the forms

\[ \rho = \rho(\theta, p^2 \theta, (p \theta)^2, \ldots) = \sum_{n=0}^{\infty} e^{i2n} \rho_n. \]

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\begin{equation}
\dot{\theta} = \Omega(\theta, \theta^2, (\theta^0)^2, \cdots) = \sum_{n=0}^{\infty} \varepsilon^n \Omega_n.
\end{equation}

The zeroth order terms \( \rho_0 \) and \( \Omega_0 \) should have functional forms identical with (3) and (4), respectively. Since the appearance of \( \varepsilon \) yields a small quantity of order \( \varepsilon^2 \), the \( \varepsilon^2 \)-correction terms should be of the forms
\begin{align}
\rho_1 &= \alpha \theta^2 + \beta (\theta^0)^2, \\
\Omega_1 &= \varepsilon \theta^2 + \mu (\theta^0)^2,
\end{align}
where \( \alpha, \beta, \nu \) and \( \mu \) are generally dependent on \( \theta \). Requiring that the original equations of motion should identically be satisfied by the expansion forms (6), one may determine \( \alpha, \beta, \nu \) and \( \mu \). A calculation shows that \( \alpha \) and \( \beta \) should satisfy certain first order linear differential equations, which we write
\begin{equation}
\frac{d\alpha}{d\theta} = p(\theta) \alpha + q(\theta)
\end{equation}
and similarly for \( \beta \). On the other hand, \( \nu \) and \( \mu \) are simply given by some linear functions of \( \alpha \) and \( \beta \), respectively. The \( \theta \)-dependence of \( p \) and \( q \) comes from their dependence on \( \rho_0 \) and \( \Omega_0 \) as well as on \( \sin \theta \) and \( \cos \theta \). Thus \( p \) and \( q \) practically become \( 2\pi \)-periodic in \( \theta \). From this fact and the fact that the limit cycle represented by Eq. (2) is a stable one, one may conclude that the solution of (8) reduces to a definite \( 2\pi \)-periodic function of \( \theta \) for \( t \to \infty \) (viz., \( \theta \to +\infty \) or \( -\infty \)). A similar argument shows that \( \beta, \nu \) and \( \mu \) also become \( 2\pi \)-periodic in \( \theta \) for \( t \to \infty \). The replacement of all the relevant \( \theta \)-dependent quantities by their asymptotic forms for \( t \to \infty \) is justified whenever the aforementioned initial period is much shorter than the time scale of order \( \varepsilon^{-2} \), a new time scale appearing when the spatial variation of \( \theta \) is included. In this way one finally obtains
\begin{equation}
\dot{\theta} = \Omega_\theta(\theta, \theta^2, (\theta^0)^2, \cdots) = \sum_{n=0}^{\infty} \varepsilon^n \Omega_n.
\end{equation}

Even if the last two terms in Eq. (9) are much smaller than \( \Omega_0 \), the former cannot be neglected because the difference in phases of sufficiently distant points in space cannot necessarily be small. This fact also implies that the phase itself should not be expanded in powers of \( \varepsilon \). On the contrary, the amplitude \( \rho \) may be approximated by \( \rho_0 \) for slow spatial variation without affecting the concentration patterns significantly. Thus Eq. (5) may still be retained, while the equation for \( \theta \) should be changed from (4) to (9).

Equations (5) and (9) constitute a model suitable for investigating the pattern formation in a reaction-diffusion system. Specifically, concentration "isobar" \( r=r(t) \) is simply given by
\begin{equation}
\theta(\mathbf{r}, t) + \theta_0 = 2n\pi
\end{equation}
with the solution \( \theta(\mathbf{r}, t) \) of Eq. (9): It should be noted that one need not know the explicit functional forms of \( x_0(\theta) \) and \( \gamma_0(\theta) \).

As a particularly simplified model let the \( \theta \)-dependence of \( \Omega_0, \nu \) and \( \mu \) be dropped. Such a model was also derived from the reductive perturbation method, and was applied to some specific problems of pattern formation.

Details will be explained in a forthcoming paper.