Progress of Theoretical Physics, Vol. 63, No. 6, June 1980

Instability and Turbulence of Wavefronts in Reaction-Diffusion Systems

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(Received September 19, 1979)

The theoretical possibility of a new kind of diffusion-induced chemical turbulence is discussed. Here the nearly planar wavefront of a pulse or phase-boundary propagating through a more than one-dimensional medium is of our concern. By means of an asymptotic method an equation describing the behavior of a curved wavefront is first derived assuming that its spatial variation is slow. The method employed and the resulting equation are completely analogous to those encountered in the theory of turbulent phase waves presented earlier, although the physical situations considered are different. It turns out that if a spontaneous deformation of a planar wavefront occurs, this is immediately accompanied by turbulence provided that the system is well extended. The condition for this kind of instability is shown analytically to be fulfilled by a simple activator-inhibitor model.

§ 1. Introduction

In nonequilibrium open systems, multiple diffusion processes give rise to a number of unexpected features. A most striking example is the symmetry-breaking spatial differentiation of the Rashevsky-Turing type[1)~5)] in which the difference in the diffusion rates of the activator and inhibitor plays a decisive role in patterning. It has recently been shown that the system of diffusion-coupled limit cycle oscillators is capable of showing turbulent behavior,[6)~8)] and that multiple diffusion processes with unequal diffusion rates are again important. Another example, similar to the latter phenomenon, can be found in combustion. Apart from the instability originating from the thermal expansion of the gas,[9)] the interaction of diffusion and heat conduction processes can lead to a flame-front instability and subsequent turbulence.[10),11)] For all the above phenomena, multiple diffusion processes together with some nonequilibrium conditions result in an effectively negative diffusion rate of the principal mode. The instability and turbulence of wavefronts in chemical reactions which are treated in the present paper fall into the same class of phenomena as the above.

Unlike the Rashevsky-Turing instability, however, the instabilities in the other three cases are characterized by the existence of a "translational mode". Take for example the propagation of a wavefront in a two-dimensional reaction-diffusion medium which is supposed to be well extended in both the x and y directions. Let $X(x-vt)$ represent a propagating wave (a pulse or a phase boundary). Then $X(x-vt+\psi)$ with arbitrary spatial translation ψ should also be a possible solution of the governing equation.

The fact to be noticed is the existence of a "constant of motion" ψ. This enables us, when ψ is allowed to have a slow dependence on the lateral coordinate y, to apply an asymptotic method to finding an evolution equation for $\psi(y, t)$ in a closed form. We shall find in § 2 how this can be accomplished.

Exactly the same fact was encountered in discussing the turbulent phase waves in systems of diffusion-coupled limit cycle oscillators,[8] $\psi(y, t)$ there corresponding to the phase of the local limit cycle oscillation. Indeed the analogy between the two problems is so complete that the final equations in both cases take an identical form, which is

$$\partial_t\psi = \nu\partial_y{}^2\psi + \mu(\partial_y\psi)^2 - \lambda\partial_y{}^4\psi\,, \tag{1·1}$$

if $|\nu|$ is sufficiently small. It is interesting to note that Sivashinsky[12] also derived the same equation when studying turbulent flamefront, but using a different method from ours.

In § 4 we shall find that the quantity ν may be negative, just as in the case of phase turbulence and combustion. The behavior of ψ for negative ν has already been studied in some detail,[8),12] and turbulent behavior was obtained if the system size was sufficiently large. Thus, in the present paper, we shall concentrate on deriving an equation of the form (1·1), and then provide an example giving rise to negative ν, rather than analysing Eq. (1·1) itself except briefly in § 3. In § 5, a physical interpretation of the wavefront instability will be given.

§ 2. General formulation

Consider a two-dimensional reaction-diffusion system composed of n concentration variables $(X_1, X_2, \cdots, X_n) \equiv \boldsymbol{X}$ obeying the following equation in vector form:

$$\partial_t\boldsymbol{X} = \boldsymbol{F}(\boldsymbol{X}) + D(\partial_x{}^2 + \partial_y{}^2)\boldsymbol{X}\,, \tag{2·1}$$

where D is a diffusion matrix assumed to be diagonal, and abbreviations such as ∂_t and ∂_x in place of $\partial/\partial t$ and ∂/∂_x are used. The formulation below may readily be generalized to the three-dimensional case, and we shall not discuss its generalization in detail. Suppose that Eq. (2·1) allows a steadily propagating solution

$$\boldsymbol{X} = \boldsymbol{X}_s(x - vt) \tag{2·2}$$

in an infinite medium. In particular we will be concerned with a propagating single pulse or single phase-boundary rather than periodic wave trains. The wave patterns of our concern are shown schematically in Fig. 1. Obviously, $\boldsymbol{X}_s$ satisfies

$$\boldsymbol{F}(\boldsymbol{X}_s(z)) + (Dd_z^2 + vd_z)\boldsymbol{X}_s(z) = 0\,, \tag{2·3}$$

where z is the moving coordinate,

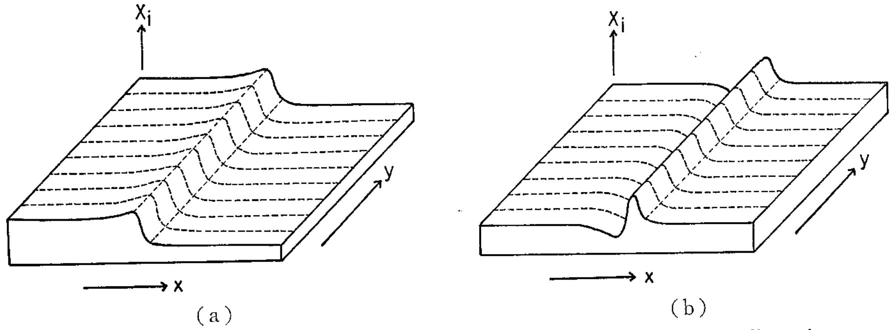


Fig. 1. Schematic wave patterns of a component X_i without front distortion.
(a) Propagating phase boundary. (b) Propagating pulse.

$$z=x-vt\,. \tag{2·4}$$

We assume that $\boldsymbol{X}_s$ represents a stable solution with respect to a small disturbance $\boldsymbol{u}(z,t)$, which does not depend on y. To be precise, we substitute the expression

$$\boldsymbol{X}(z,t)=\boldsymbol{X}_s(z)+\boldsymbol{u}(z,t)=\boldsymbol{X}_s+\sum_l e^{\lambda_l t}\boldsymbol{u}_l(z) \tag{2·5}$$

into Eq. (2·1) to obtain

$$\Gamma\boldsymbol{u}_l=\lambda_l\boldsymbol{u}_l \tag{2·6}$$

with

$$\Gamma=\Gamma_0+Dd_z^2+vd_z\,, \tag{2·7a}$$

$$(\Gamma_0)_{ij}=\partial F_i(\boldsymbol{X}_s)/\partial X_{sj}\,. \tag{2·7b}$$

The above-mentioned stability condition for $\boldsymbol{X}_s$ means that no eigenvalue λ_l should have a positive real part. However, it is important to notice that among the λ_l at least one eigenvalue is exactly zero because of the equation

$$\Gamma d_z\boldsymbol{X}_s=0\,, \tag{2·8}$$

which is obtained by the application of d_z to Eq. (2·3). By comparison of Eqs. (2·8) and (2·6), one may put

$$\boldsymbol{u}_0=d_z\boldsymbol{X}_s\,, \tag{2·9a}$$

$$\lambda_0=0\,. \tag{2·9b}$$

The existence of the zero eigenvalue is a natural consequence of the fact that any spatial translation of $\boldsymbol{X}_s(z)$ produces a solution of Eq. (2·1). In the discussion below we always assume that all the other eigenvalues have negative real parts, and that the zero eigenvalue is isolated. The latter assumption is expected to be true for the waves under consideration, but may not be true for periodic wave trains.

It is appropriate here to introduce notation. We define the adjoint operator Γ^* of Γ by

$$\int_{-\infty}^{\infty} \boldsymbol{g}(z)(\Gamma \boldsymbol{f}(z))dz = \int_{-\infty}^{\infty} (\Gamma^* \boldsymbol{g}(z))\boldsymbol{f}(z)dz \tag{2·10}$$

for arbitrary vector functions $\boldsymbol{f}(z)$ and $\boldsymbol{g}(z)$ having the property: $\boldsymbol{f}$, $\boldsymbol{g}$, $d_z\boldsymbol{f}$, $d_z\boldsymbol{g} \to 0$ as $|z| \to \infty$. It is easy to see that Γ^* is explicitly given by

$$\Gamma^* = {}^t\Gamma_0 + Dd_z^2 - vd_z , \tag{2·11}$$

where ${}^t\Gamma_0$ denotes the transpose of Γ_0. Correspondingly, we introduce the eigenvectors $\boldsymbol{u}_l^*(z)$ satisfying

$$\Gamma^* \boldsymbol{u}_l^* = \lambda_l \boldsymbol{u}_l^*, \tag{2·12}$$

together with the orthonormality condition

$$\int_{-\infty}^{\infty} {}^t\boldsymbol{u}_l^* \boldsymbol{u}_m dz = \delta_{lm} . \tag{2·13}$$

For the sake of brevity, the following notation will be used:

$$A_{lm} \equiv \int_{-\infty}^{\infty} {}^t\boldsymbol{u}_l^* A \boldsymbol{u}_m dz , \tag{2·14}$$

where the $n \times n$ matrix A may contain functions of z and/or differential operators with respect to z.

As a generalization of the solution in Eq. (2·2), we are now interested in an essentially two-dimensional solution $\boldsymbol{X}(z, y, t)$ such that its wave profile along z for any given y does not much differ from $\boldsymbol{X}_s(z-\psi)$ with an appropriate choice of the phase ψ, but ψ itself shows a slow spatial variation in the lateral direction y. Such a wave pattern is schemetically shown in Fig. 2. However, the wave pattern of $\boldsymbol{X}(z, y, t)$ cannot be made identical with $\boldsymbol{X}_s(z-\psi)$ by any choice of $\psi(y)$, so that we have generally to put

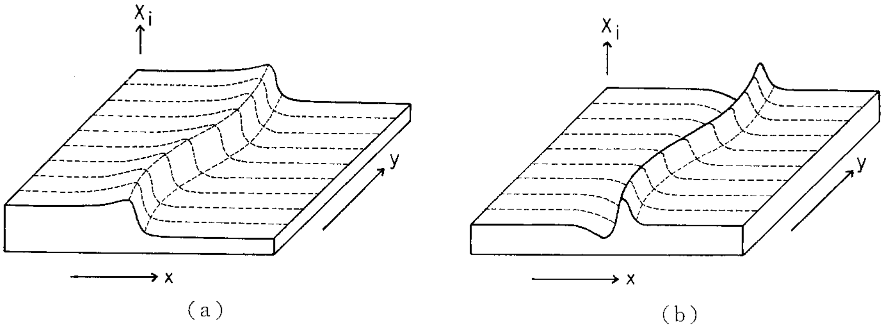


Fig. 2. Schematic wave patterns of a component X_i with slow lateral variation of the fronts. (a) Propagating phase boundary. (b) Propagating pulse.

$$\boldsymbol{X}(z, y, t) = \boldsymbol{X}_s(z-\psi(y, t)) + \boldsymbol{u}(z-\psi(y, t), y, t), \tag{2·15}$$

where $\boldsymbol{u}$ takes account of the uncompensated deformation of the wave profile. We have now to inquire into the meaning of the phrase "appropriate choice of ψ". The most natural definition of ψ will be such that any translational disturbance is excluded from $\boldsymbol{u}$, namely,

$$\int_{-\infty}^{\infty} {}^t\boldsymbol{u}_0^*(z)\,\boldsymbol{u}(z, y, t)\,dz = 0\,. \tag{2·16}$$

Besides making the decomposition (2·15) unique, the above orthogonality condition has an important physical implication. The absence of the translational mode in $\boldsymbol{u}$ implies that it describes only the rapid adiabatic processes which follow the slow process described by $\boldsymbol{X}_s(z-\psi)$. At such a dynamical stage, a great reduction of the dynamics may be accomplished by a *functional postulate.* We now apply this postulation to the pair of small quantities $\boldsymbol{u}$ and $\partial_t\psi$ assuming them to be of the form

$$\boldsymbol{u}(z-\psi(y, t), y, t) = \boldsymbol{u}(z-\psi(y, t), \partial_y^2\psi, (\partial_y\psi)^2, \partial_y^4\psi, \cdots), \tag{2·17a}$$

$$\partial_t\psi = \Omega(\partial_y^2\psi, (\partial_y\psi)^2, \partial_y^4\psi, \cdots). \tag{2·17b}$$

Note that the above expressions do not contain terms like $\partial_y\psi, \partial_y^3\psi, \partial_y^2\psi\partial_y\psi, \cdots$, that is, terms containing odd numbers of spatial derivatives. We have assumed this property so that dynamically reduced equations (2·17) may preserve the invariance under the spatial inversion $y\to -y$, the property obviously possessed by the original equation (2·1).

We note that in the absence of the spatial variation of ψ, the quantities $\boldsymbol{u}$ and $\partial_t\psi$ should vanish, recovering the solution (2·2). It is therefore appropriate to make a perturbation expansion of $\boldsymbol{u}$ and $\partial_t\psi$ in powers of ∂_y. A systematic way for doing this is to introduce an indicator of smallness ε, and make the replacement

$$\partial_y \to \varepsilon\partial_y \tag{2·18}$$

in Eq. (2·17). Then we expand $\boldsymbol{u}$ and $\partial_t\psi$ in powers of ε, and finally put $\varepsilon=1$. The expansion forms will generally be

$$\begin{aligned}\boldsymbol{u}(z', y, t) &= \varepsilon^2\{\boldsymbol{u}_1^{(1)}(z')\,\partial_y^2\psi + \boldsymbol{u}_1^{(2)}(z')\,(\partial_y\psi)^2\} \\ &\quad + \varepsilon^4\{\boldsymbol{u}_2^{(1)}(z')\,\partial_y^4\psi + \cdots\} + O(\varepsilon^6),\end{aligned} \tag{2·19a}$$

$$\partial_t\psi = \varepsilon^2\{\Omega_1^{(1)}\partial_y^2\psi + \Omega_1^{(2)}(\partial_y\psi)^2\} + \varepsilon^4\{\Omega_2^{(1)}\partial_y^4\psi + \cdots\} + O(\varepsilon^6), \tag{2·19b}$$

where

$$z' = z - \psi(y, t), \tag{2·20}$$

and the quantities $\boldsymbol{u}_\alpha^{(\beta)}(z')$ and $\Omega_\alpha^{(\beta)}$ are as yet unspecified. It can be seen that, provided that all the $\Omega_\alpha^{(\beta)}$ are known, Eq. (2·19b) is the evolution equation, in closed form, for the phase function ψ. Equation (2·19a) has no direct importance in the present theory.

The condition (2·16) now takes the form

$$\int_{-\infty}^{\infty} {}^t\boldsymbol{u}_0^*(z)\,\boldsymbol{u}_\alpha^{(\beta)}(z)\,dz=0\,, \tag{2·21}$$

or, equivalently,

$$\boldsymbol{u}_\alpha^{(\beta)}(z)=\sum_{l\neq 0} C_{\alpha l}^{(\beta)}\boldsymbol{u}_l(z)\,. \tag{2·22}$$

On substituting Eq. (2·15) together with Eqs. (2·19a) and (2·19b) into Eq. (2·1), one may determine the quantities $\boldsymbol{u}_\alpha^{(\beta)}$ and $\Omega_\alpha^{(\beta)}$ with the aid of the condition (2·21). The explicit procedure is given in Appendix A. In particular, after putting $\varepsilon=1$, Eq. (2·19b) takes the form

$$\partial_t\psi=\nu\partial_y^2\psi+\mu(\partial_y\psi)^2-\lambda\partial_y^4\psi+\cdots \tag{2·23}$$

with the coefficients

$$\nu=D_{00}\,, \tag{2·24a}$$

$$\mu=-(Dd_z)_{00}\,, \tag{2·24b}$$

$$\lambda=\sum_{l\neq 0}\lambda_l^{-1}D_{0l}D_{l0}\,, \tag{2·24c}$$

$$\cdots\cdots\cdots,$$

where the notation defined by Eq. (2·14) has been used. Equation (2·23) is the basic equation describing the evolution of the wavefront.

§ 3. Cases of interest

In this section we consider two special cases for which Eq. (2·23) turns out to be useful. If the spatial variation of ψ is sufficiently slow, and if all the coefficients $\nu, \mu, \lambda\cdots$ are quantities of normal magnitudes, then in Eq. (2·23) only the lowest order terms in ∂_y need be retained, to give

$$\partial_t\psi=\nu\partial_y^2\psi+\mu(\partial_y\psi)^2. \tag{3·1}$$

We assume that ν is positive, otherwise this approximate equation can be shown to be meaningless. Exactly the same equation has been derived and used in discussing pattern formation in systems of diffusion-coupled limit cycle oscillators.[13] However, the peculiarity of the present problem is that the nonlinear term $\mu(\partial_y\psi)^2$ can be interpreted physically in the following simple manner. As is proved in Appendix B, we have in general the identity

$$\mu=v/2\,. \tag{3·2}$$

On the other hand, the propagation velocity in the z direction is obviously $v + \partial_t\psi$, and this quantity should in general be related to the propagation velocity v_ξ normal to the wavefront by the equation

$$v+\partial_t\psi = v_\xi\sqrt{1+(\partial_y\psi)^2}. \tag{3·3}$$

The reason will be clear from Fig. 3. Since $\partial_t\psi$ and $(\partial_y\psi)^2$ are small compared with v, Eq. (3·3) may be approximated by

$$v+\partial_t\psi = v_\xi + \frac{v}{2}(\partial_y\psi)^2. \tag{3·4}$$

Comparing the above with Eq. (3·1) taking account of Eq. (3·2), we find

$$|v_\xi| = |v| - \nu\kappa\,. \tag{3·5}$$

Here κ is the curvature of the front, and is taken to be positive if the front is convex and negative if concave. Thus Eq. (3·5) simply means that the propagation speed normal to the front is modified only by a curvature effect. In particular, if ν is positive, the front moves so as to make itself smoother, just as the surface tension at the liquid-vapor interface has the same effect. If the front is concave, the above smoothening effect is balanced with the sharpening effect due to the wave propagation, and one may expect the appearance of a shock structure. Equation (3·1) indeed admits a family of shock solutions

$$\psi = \frac{v}{2}(a^2+b^2)t + ay + \frac{2\nu}{v}\ln\left[\cosh\left(\frac{bv}{2\nu}(y+avt)\right)\right], \tag{3·6}$$

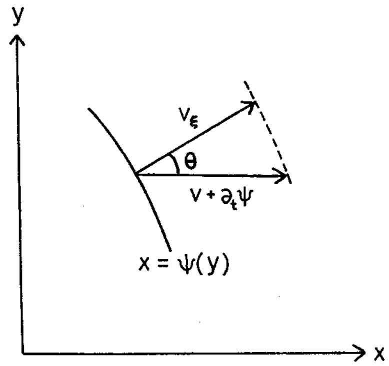


Fig. 3. Relation of $\partial_t\psi$ to the propagation velocity v_ξ normal to the wavefront. The x-coordinate of the front is $\psi(y)$, which implies the relation $\cos\theta = 1/\sqrt{\{1+(\partial_y\psi)^2\}}$ giving Eq. (3·3).

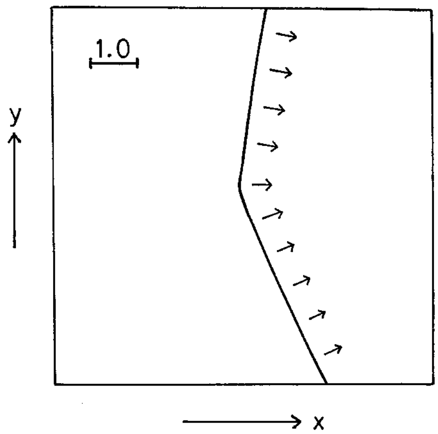


Fig. 4. A shock pattern corresponding to Eq. (3·6). Parameter values: $a=-0.15$, $b=0.30$, $\nu=1.0$ $v=10.0$.

where a and b are parameters related to the slope of the wavefront at infinity by the equation

$$\lim_{y\to\pm\infty} \partial_y\psi = a \pm b\,. \tag{3·7}$$

The front pattern described by Eq. (3·6) is illustrated in Fig. 4. It is interesting to note that Eq. (3·1) is transformed into the Burgers equation[13]

$$\partial_t U = \nu\partial_y^2 U + 2\mu U\partial_y U \tag{3·8}$$

through $U = \partial_y\psi$, so that the solution (3·6) is mathematically equivalent to the well-known shock solution of the Burgers equation.

Another interesting feature is provided by negative ν, the case of "negative surface tension". Equation (3·1) or (3·8) then becomes meaningless, because the latter can be transformed into the diffusion equation

$$\partial_t f = \nu\partial_y^2 f \tag{3·9}$$

with the negative diffusion constant through the Hopf-Cole transformation

$$f = \exp\left[\mu\nu^{-1}\int U dy\right]. \tag{3·10}$$

Thus we must necessarily take account of some higher order terms in Eq. (2·23). Suppose that $|\nu|$ is still small. Then the most important higher order term can be shown to be $\partial_y^4\psi$, and the resulting equation is

$$\partial_t\psi = \nu\partial_y^2\psi + \mu(\partial_y\psi)^2 - \lambda\partial_y^4\psi\,, \tag{3·11}$$

where $\lambda > 0$ is assumed. In fact the solution of the above equation has the scaling form

$$\psi = |\nu|\tilde{\psi}(|\nu|^{1/2}y, \nu^2 t)\,, \tag{3·12}$$

making all the other terms, excluded from Eq. (3·11), higher order in ν. In previous papers[6),8)] it was shown that the solution of Eq. (3·11) is turbulent if the system size is sufficiently large. Therefore a discussion on the behavior of ψ will not be repeated. Instead we turn now to finding an example producing negative ν.

§ 4. A soluble example showing negative ν

The example we will consider in this section is a piecewise linear version of the Bonhoeffer-van der Pol model with diffusion. Our purpose here is only to demonstrate the occurrence of negative ν. For the calculation of ν it suffices to consider a one-dimensional system. The equations considered are

$$\partial_t X = -X - Y + H(X-a) + D_X \partial_x^2 X, \tag{4·1a}$$

$$\partial_t Y = bX - cY + D_Y \partial_x^2 Y. \tag{4·1b}$$

Here H is the Heaviside step function, and the parameters a, b and c are assumed to be in the range

$$0 < a < \tfrac{1}{2}, \qquad b, c > 0. \tag{4·2}$$

This kind of model has been studied by several people. Rinzel and Keller[14] discussed analytically traveling pulses and their stability for the case $c = D_Y = 0$; Winfree[15] obtained by computer calculation a two-dimensional spiral pattern for the case $c=0$, $D_X = D_Y$; Koga and Kuramoto[16] recently demonstrated the existence of some propagationless solitary patterns for the case $D_Y \gg D_X$.

For the diffusionless case, our system has the following properties. There is always a stable fixed point at $(X, Y) = (0, 0)$. If the condition $c/(b+c) > a$ is satisfied, there is also another stable fixed point (X_0, Y_0), where

$$X_0 = \frac{c}{b+c}, \quad Y_0 = \frac{b}{b+c}. \tag{4·3}$$

We shall retain this notation for X_0 and Y_0 even if the bistability condition is not satisfied. The nullcline $d_t X = 0$ forms a sigmoidal manifold. Thus X forms a hysterisis subsystem containing Y as a hysterisis eliciting parameter. It may also be said that X is an activating substance, Y being an inhibiting one.

Our present concern is a steadily propagating solution

$$X = X_s(z), \qquad Y = Y_s(z) \tag{4·4}$$

with $z = x - vt$. The boundary condition

$$X_s(\infty) = Y_s(\infty) = 0 \tag{4·5}$$

is always assumed. In particular we concentrate on the following two types of solutions.

(A) A single front propagation for the bistable case with the boundary conditions

$$X_s(-\infty) = X_0, \qquad Y_s(-\infty) = Y_0, \tag{4·6a}$$

$$X_s(0) = a. \tag{4·6b}$$

(B) A single pulse propagation for the monostable and bistable cases with the boundary conditions

$$X_s(-\infty) = Y_s(-\infty) = 0, \tag{4·7a}$$

$$X_s(\sigma) = X_s(0) = a, \tag{4·7b}$$

where σ is, as yet, an unspecified pulse width.

The method of finding such solutions is almost identical to that of Rinzel and Keller, so that we omit all the calculational details. Thanks to the piecewise linear character of the model, in both cases the solution may be expressed in terms of exponential functions $\exp(\alpha_i z)$. Here α_i are the roots of the equation

$$P(\alpha)=f(\alpha)g(\alpha)+b=0\,, \tag{4·8}$$

where

$$f(\alpha)=D_X\alpha^2+v\alpha-1\,, \tag{4·9a}$$

$$g(\alpha)=D_Y\alpha^2+v\alpha-c\,. \tag{4·9b}$$

The coefficients before the exponential functions are determined from the boundary conditions, and also from the continuity conditions for X_s, Y_s and their first spatial derivatives at the junction points. Further, condition (4·6b) is used for the determination of v and similarly the condition (4·7b), for v and σ. The solution (X_s, Y_s) thus obtained is given in Appendix C.

The next thing to do is to find the null eigenvectors

$$\boldsymbol{u}_0^{(z)}\equiv\begin{pmatrix}x_0(z)\\ y_0(z)\end{pmatrix},\quad \boldsymbol{u}_0^*\equiv\begin{pmatrix}x_0^*(z)\\ y_0^*(z)\end{pmatrix} \tag{4·10}$$

of the operators Γ and Γ^*, respectively, with which the quantity ν may be expressed as

$$\nu=D_{00}=(G_XD_X+G_YD_Y)/(G_X+G_Y)\,, \tag{4·11}$$

where

$$G_X=\int_{-\infty}^{\infty}x_0^*(z)x_0(z)dz\,, \tag{4·12a}$$

$$G_Y=\int_{-\infty}^{\infty}y_0^*(z)y_0(z)dz\,. \tag{4·12b}$$

If $\boldsymbol{u}_0$ and $\boldsymbol{u}_0^*$ are defined as normalized quantities, we have of course $G_X+G_Y=1$. One may recall here that $\boldsymbol{u}_0$ is simply given by the first derivative of the above-obtained propagating solutions according to Eq. (2·9a). Thus the only remaining problem is to find $\boldsymbol{u}_0^*(z)$. The method for obtaining $\boldsymbol{u}_0^*$, and the expressions for $\boldsymbol{u}_0^*$, G_X and G_Y is given in Appendix D.

As is implied by Eqs. (D·12) and (D·13), the expression for ν is quite complicated. It is possible, with the aid of a numerical procedure, to examine the sign of ν for various parameter values. In order to continue the study analytically, however, the problem is simplified (but not made trivial) by allowing some of the parameters to have extremely small values. In particular, we are interested in the case

$$D_X=\varepsilon^{1/2}\tilde{D}_X,\qquad b=\varepsilon\tilde{b},\qquad c=\varepsilon\tilde{c}\,, \tag{4·13}$$

where ε is a small quantity, while $\tilde{D}_X$, $\tilde{b}$, $\tilde{c}$ as well as the other parameters are assumed to have normal magnitudes. In the limit $\varepsilon \to 0$, the expressions for various quantities which we are concerned with are greatly simplified as follows. Assuming that the propagation velocity v is positive and of the order of $\varepsilon^{1/4}$, which is actually the case as will be seen presently, the four roots of Eq. (4·8) are reduced to

$$\alpha_{1,3} = \frac{\varepsilon^{-1/4}}{2\tilde{D}_X}(-\tilde{v} \pm \sqrt{\tilde{v}^2 + 4\tilde{D}_X}), \tag{4·14a}$$

$$\alpha_2 = \varepsilon^{3/4}\tilde{v}^{-1}(\tilde{b} + \tilde{c}), \tag{4·14b}$$

$$\alpha_4 = -\varepsilon^{1/4}\tilde{v}D_Y^{-1}, \tag{4·14c}$$

where $\tilde{v}$ is the scaled velocity

$$\tilde{v} = \varepsilon^{-1/4}v\,. \tag{4·15}$$

In terms of the scaled coordinate ξ defined by

$$\xi = \varepsilon^{3/4}z\,, \tag{4·16}$$

and of the scaled quantities

$$\tilde{\sigma} = \varepsilon^{1/4}\sigma\,, \tag{4·17a}$$

$$\tilde{\alpha} = \varepsilon^{-1/4}\alpha_2 = \tilde{v}^{-1}(\tilde{b} + \tilde{c})\,, \tag{4·17b}$$

our propagating solutions in Eqs. (C·1) and (C·3) are reduced to
(A)

$$X_s(\xi) = X_0 + Y_0 \exp(\tilde{\alpha}\xi)\,, \qquad \xi < 0 \tag{4·18a}$$

$$= 0\,, \qquad \xi > 0 \tag{4·18b}$$

$$Y_s(\xi) = Y_0(1 - \exp(\tilde{\alpha}\xi))\,, \qquad \xi < 0 \tag{4·18c}$$

$$= 0\,. \qquad \xi > 0 \tag{4·18d}$$

(B)

$$X_s(\xi) = Y_0(\exp(-\tilde{\alpha}\tilde{\sigma}) - 1)\exp(\tilde{\alpha}\xi)\,, \qquad \xi < 0 \tag{4·19a}$$

$$= X_0 + Y_0 \exp[\tilde{\alpha}(\xi - \tilde{\sigma})]\,, \qquad 0 < \xi < \tilde{\sigma} \tag{4·19b}$$

$$= 0\,, \qquad \xi > \tilde{\sigma} \tag{4·19c}$$

$$Y_s(\xi) = Y_0(1 - \exp(-\tilde{\alpha}\tilde{\sigma}))\exp(\tilde{\alpha}\xi)\,, \qquad \xi < 0 \tag{4·19d}$$

$$= Y_0(1 - \exp[\tilde{\alpha}(\xi - \tilde{\sigma})])\,, \qquad 0 < \xi < \tilde{\sigma} \tag{4·19e}$$

$$= 0\,. \qquad \xi > \tilde{\sigma} \tag{4·19f}$$

In both Cases (A) and (B) the scaled propagation velocity is given by

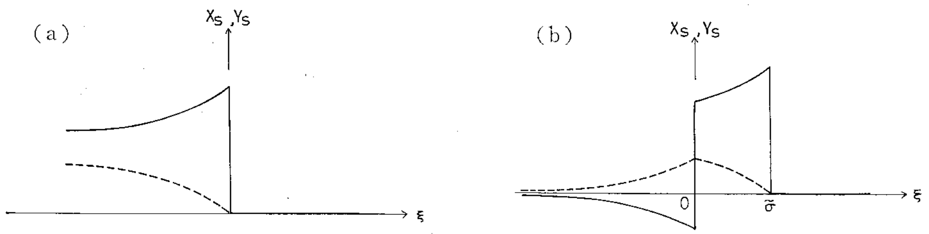


Fig. 5. Analytically obtained wave patterns for the extreme case (4·13); (a) and (b) correspond to Eqs. (4·18) and (4·19), respectively. Solid curves represent X_s, and broken curves Y_s.

$$\tilde{v}=(1-2a)\sqrt{\frac{\tilde{D}_X}{a(1-a)}}\,, \tag{4·20}$$

and the scaled pulse width for case (B) is

$$\tilde{c}=\frac{\tilde{v}}{\tilde{b}+\tilde{c}}\ln\frac{Y_0}{2a-X_0}\,. \tag{4·21}$$

The wave patterns corresponding to Eq. (4·18) and (4·19) are shown in Fig. 5. The expression for ν obtained from Eqs. (D·12) and (4·11) is also greatly simplified, and turns out to be identical for Cases (A) and (B). We have from Eqs. (D·12a) and (D·12b)

$$G_X=\frac{\tilde{D}_X}{\varepsilon^{1/4}(\tilde{v}^2+4\tilde{D}_X)^{3/2}}\,, \tag{4·22a}$$

$$G_Y=-\varepsilon^{1/4}bD_Y/\tilde{v}^3, \tag{4·22b}$$

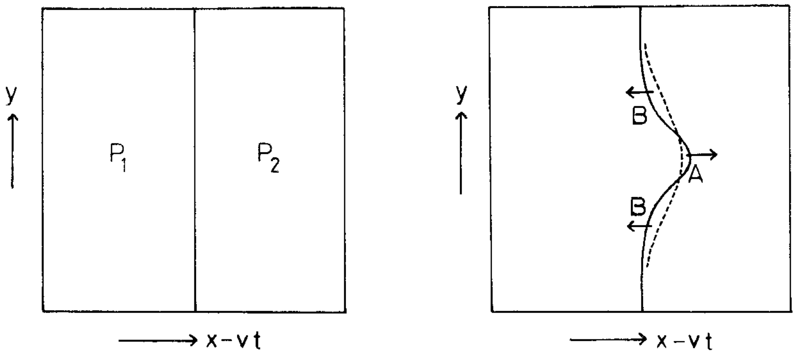
so that

$$\nu=\varepsilon^{1/2}\tilde{D}_X\left\{1-\frac{\tilde{b}}{(1-2a)^2}\left(\frac{D_Y}{\tilde{D}_X}\right)^2\right\}. \tag{4·23}$$

Note that we have $|G_Y/G_X|\to 0$ as $\varepsilon\to 0$, still the quantities G_XD_X and G_YD_Y have the same order of magnitude. Thus the transition from positive to negative ν is possible by changing some parameter, e.g., $\tilde{D}_X$. For any scaling choice different from that in Eq. (4·13) such a transition is impossible. For instance, if we take $D_X=\varepsilon\tilde{D}_X$ keeping b and c as in Eq. (4·13), the quantity ν turns out to be definitely negative.

§ 5. Discussion

Here we supplement the results of the preceding sections with some qualitative arguments. The reason for the occurrence of the spontaneous deformation of a wavefront may be qualitatively understood as follows. In order to make the arguments



(a) (b)

Fig. 6. Instability of a wavefront. For explanation, see text.

concrete, consider a bistable system composed of an activator X and inhibitor Y. Suppose that the medium is now partitioned into two subregions, each corresponding to a uniform steady state $P_1=(X_1, Y_1)$ or $P_2=(X_2, Y_2)$ as shown in Fig. 6(a). The boundary separating these regions is a straight line, and it propagates along the x direction. Thus the region P_1 will be the activated region, namely, $X_1>X_2$, so that Y_1 should also be larger than Y_2. A part of the wavefront is now pushed forward as in Fig. 6(b), and we ask what happens subsequently. The fact that the diffusion constants D_X and D_Y are positive implies that the phase boundary possesses a kind of surface tension acting as a flattening force on the front non-uniformity. If D_Y is much greater than D_X, however, there exists a sizable counter-effect. In fact, in such a case, Y will diffuse rapidly out of the promontory A as indicated by the dotted line in Fig. 6(b), and this will bring about the scarcity of Y in A. Thus the autocatalytic production of X is accelerated, and the propagation speed is increased at the part A. What happens in the neighbourhood B is the exact converse. Namely, Y is excessive there so that the propagation speed will be diminished. As a result, the shape of the boundary tends to be distorted further. If such a destabilization force dominates the stabilizing force mentioned at first, we have an effectively negative surface tension. Then, the boundary as a straight line becomes unstable, and this should be accompanied by the appearance of some new structure.

From a mathematical point of view, the negative sign of ν in the example considered in § 4 comes from the difference in the signs of G_X and G_Y. This can be further traced back to the existence of an antisymmetric part in the matrix Γ. Such a property of Γ is characteristic of an activator-inhibitor system. In addition, the condition $D_Y \gg D_X$ makes the negative contribution to ν greater as is seen from Eq. (4·11). Such mathematical reasoning is in accordance with the intuitive picture described in the preceding paragraph.

Just as the occurrence of a propagationless solitary pattern discussed in the previous paper,[16] the instability and turbulence of wavefronts might be considered to be one of the phenomena resulting from the competition between the cross-inhibitory and cross-excitory natures of the system. If one compares Eq. (4·13) with Eq. (3·1) of Ref. 16), it can be seen that the latter requires stronger cross-inhibition than the former. Namely, in the present phenomenon, the cross-inhibition is not strong enough to realize pattern localization, still it may be strong enough to give rise to a spontaneous deformation of the front and hence turbulence.

Appendix A

We explain here how the quantities $\boldsymbol{u}_\alpha^{(\beta)}$ and $\Omega_\alpha^{(\beta)}$ appearing in Eq. (2·19) are determined from Eqs. (2·1) and (2·19). On substituting Eq. (2·19) into Eq. (2·1), the three terms constituting Eq. (2·1) may be expressed as

$$\begin{aligned}\partial_t \boldsymbol{X} &= \partial_t \boldsymbol{X}_s + \partial_t \boldsymbol{u} = -(v+\partial_t\psi)\, d_{z'}X_s + \partial_t \boldsymbol{u}\\ &= -v\partial_{z'}\boldsymbol{X}_s - \{\varepsilon^2(\Omega_1^{(1)}\partial_y^2\psi + \Omega_1^{(2)}(\partial_y\psi)^2) + \varepsilon^4(\Omega_2^{(1)}\partial_y^4\psi + \cdots) + \cdots\}\,\partial_{z'}\boldsymbol{X}_s\\ &\quad -\varepsilon^2 v(d_{z'}\boldsymbol{u}_1^{(1)}\partial_y^2\psi + d_{z'}\boldsymbol{u}_1^{(2)}(\partial_y\psi)^2) + \varepsilon^4(-vd_{z'}\boldsymbol{u}_2^{(1)} + \boldsymbol{u}_1^{(1)}\Omega_1^{(1)})\,\partial_y^4\psi\\ &\quad + \cdots, \end{aligned} \tag{A·1}$$

$$\boldsymbol{F}(\boldsymbol{X}) = \boldsymbol{F}(\boldsymbol{X}_s) + \Gamma\{\varepsilon^2(\boldsymbol{u}_1^{(1)}\partial_y^2\psi + \boldsymbol{u}_1^{(2)}(\partial_y\psi)^2) + \varepsilon^4(\boldsymbol{u}_2^{(1)}\partial_y^4\psi + \cdots) + \cdots\} + \cdots, \tag{A·2}$$

$$\begin{aligned} D(\partial_x^2+\partial_y^2)\,\boldsymbol{X} &= D(\partial_z^2+\partial_y^2)\,(\boldsymbol{X}_s+\boldsymbol{u})\\ &= Dd_{z'}^2\boldsymbol{X}_s - \varepsilon^2 D(d_{z'}\boldsymbol{X}_s\partial_y^2\psi - d_{z'}^2\boldsymbol{X}_s(\partial_y\psi)^2)\\ &\quad + \varepsilon^2 D(d_{z'}^2\boldsymbol{u}_1^{(1)}\partial_y^2\psi + d_{z'}^2\boldsymbol{u}_1^{(2)}(\partial_y\psi)^2) + \varepsilon^4 D(d_{z'}^2\boldsymbol{u}_2^{(1)} + \boldsymbol{u}_1^{(1)})\,\partial_y^4\psi + \cdots. \end{aligned} \tag{A·3}$$

In the above, all terms with regard to the types $\partial_y^2\psi$, $(\partial_y\psi)^2$ and $\partial_y^4\psi$ have explicitly been written down. Equation (2·1) may now be written in the form

$$\begin{aligned} &\boldsymbol{f}_0^{(1)}(z') + \varepsilon^2(\boldsymbol{f}_1^{(1)}(z')\,\partial_y^2\psi + \boldsymbol{f}_1^{(2)}(z')\,(\partial_y\psi)^2)\\ &\qquad + \varepsilon^4(\boldsymbol{f}_2^{(1)}(z')\,\partial_y^4\psi + \cdots) + O(\varepsilon^6) = 0, \end{aligned} \tag{A·4}$$

where

$$\boldsymbol{f}_0^{(1)} = \boldsymbol{F}(\boldsymbol{X}_s) + (Dd_{z'}^2 + vd_{z'})\,\boldsymbol{X}_s, \tag{A·5a}$$

$$\boldsymbol{f}_1^{(1)} = (\Omega_1^{(1)} - D)\,\boldsymbol{u}_0 + \Gamma\boldsymbol{u}_1^{(1)}, \tag{A·5b}$$

$$\boldsymbol{f}_1^{(2)} = (\Omega_1^{(2)} + Dd_{z'})\,\boldsymbol{u}_0 + \Gamma\boldsymbol{u}_1^{(2)}, \tag{A·5c}$$

$$\boldsymbol{f}_2^{(1)} = \Omega_2^{(1)}\boldsymbol{u}_0 + D\boldsymbol{u}_1^{(1)} + \Gamma\boldsymbol{u}_2^{(1)} - \boldsymbol{u}_1^{(1)}\Omega_1^{(1)}, \tag{A·5d}$$

and Eq. (2·9a) has been used. In order that Eq. (A·4) holds identically, it is necessary that all the coefficients $\boldsymbol{f}_\alpha^{(\beta)}$ are identically zero. The equation $\boldsymbol{f}_0^{(1)}=0$ is automatically satisfied according to Eq. (2·3). By applying ${}^t\boldsymbol{u}_l^*$ to the equations $\boldsymbol{f}_1^{(1),(2)}=0$ from the left and integrating over dz' from $-\infty$ to $+\infty$, we obtain

$$\Omega_1^{(1)} = D_{00}\,, \tag{A·6a}$$

$$\Omega_1^{(2)} = -(Dd_z)_{00}\,, \tag{A·6b}$$

$$C_{1l}^{(1)} = \lambda_l^{-1} D_{l0}\,, \tag{A·6c}$$

$$C_{1l}^{(2)} = -\lambda_l^{-1}(Dd_z)_{00}\,, \tag{A·6d}$$

where we have used Eqs. (2·6), (2·13) and (2·22). In a similar way, we find from the equation $\boldsymbol{f}_2^{(1)}=0$,

$$\Omega_2^{(1)} = -\sum_{l\neq 0} \lambda_l^{-1} D_{0l} D_{l0}\,. \tag{A·7}$$

On putting $\Omega_1^{(1)}=\nu$, $\Omega_1^{(2)}=\mu$ and $\Omega_2^{(1)}=-\lambda$, we obtain Eqs. (2·23) and (2·24).

Appendix B

Here we give the proof of Eq. (3·2). The null eigenvectors $\boldsymbol{u}_0(z)$ and $\boldsymbol{u}_0^*(z)$ have been shown in § 2 to satisfy

$$(\Gamma_0(z) + Dd_z^2 + vd_z)\,\boldsymbol{u}_0(z) = 0\,, \tag{B·1}$$

and

$$({}^t\Gamma_0(z) + Dd_z^2 - vd_z)\,\boldsymbol{u}_0^*(z) = 0\,, \tag{B·2}$$

or, taking the transpose of Eq. (B·2),

$${}^t\boldsymbol{u}_0^*\Gamma_0(z) + d_z^2\,{}^t\boldsymbol{u}_0^*D - vd_z\,{}^t\boldsymbol{u}_0^* = 0\,. \tag{B·3}$$

We now subtract the product formed by left multiplication of (B·1) by ${}^t\boldsymbol{u}_0^*$ from the product formed by right multiplication of (B·3) by $\boldsymbol{u}_0$ to obtain

$$(d_z^2\,{}^t\boldsymbol{u}_0^*D\boldsymbol{u}_0 - {}^t\boldsymbol{u}_0^*Dd_z^2\boldsymbol{u}_0) - v(d_z\,{}^t\boldsymbol{u}_0^*\boldsymbol{u}_0 + {}^t\boldsymbol{u}_0^*d_z\boldsymbol{u}_0) = 0\,. \tag{B·4}$$

The above may be rewritten as

$$d_z\{(d_z\,{}^t\boldsymbol{u}_0^*D\boldsymbol{u}_0 - {}^t\boldsymbol{u}_0^*Dd_z\boldsymbol{u}_0) - v\,{}^t\boldsymbol{u}_0^*\boldsymbol{u}_0\} = 0\,. \tag{B·5}$$

We assume that $\boldsymbol{u}_0$ and $\boldsymbol{u}_0^*$ go to zero sufficiently rapidly as $|z|\to\infty$. Thus, Eq. (B·5) may be integrated to give

$$d_z\,{}^t\boldsymbol{u}_0^*D\boldsymbol{u}_0 - {}^t\boldsymbol{u}_0^*Dd_z\boldsymbol{u}_0 = v\,{}^t\boldsymbol{u}_0^*\boldsymbol{u}_0\,. \tag{B·6}$$

Integrating Eq. (B·6) again, using partial integration, we obtain

$$\begin{aligned} v &= \int_{-\infty}^{\infty} (d_z{}^t\boldsymbol{u}_0^*D\boldsymbol{u}_0 - {}^t\boldsymbol{u}_0^*Dd_z\boldsymbol{u}_0)\,dz \\ &= -2\int_{-\infty}^{\infty} {}^t\boldsymbol{u}_0^*Dd_z\boldsymbol{u}_0 dz = -2(Dd_z)_{00} = 2\mu\,, \end{aligned} \tag{B·7}$$

which completes the proof.

Appendix C

Assuming that Eq. (4·8) has two positive roots α_1 and α_2, and two negative ones α_3 and α_4, our steadily propagating solutions for Cases (A) and (B) may be expressed as follows:

(A)

$$X_s(z) = -\frac{g_1}{\alpha_1 Q_1}\exp(\alpha_1 z) - \frac{g_2}{\alpha_2 Q_2}\exp(\alpha_2 z) + X_0\,, \quad z \leq 0 \tag{C·1a}$$

$$= \frac{g_3}{\alpha_3 Q_3}\exp(\alpha_3 z) + \frac{g_4}{\alpha_4 Q_4}\exp(\alpha_4 z)\,, \quad z > 0 \tag{C·1b}$$

$$Y_s(z) = -X_s(z) + H(-z) + D_X d_z^2 X_s + v d_z X_s\,, \quad z \lesseqgtr 0 \tag{C·1c}$$

where

$$Q_\nu = \frac{dP(\alpha_\nu)}{d\alpha_\nu}\,, \tag{C·2a}$$

$$g_\nu = g(\alpha_\nu)\,. \tag{C·2b}$$

(B)

$$X_s(z) = -\frac{g_1}{\alpha_1 Q_1}(\mu_1^{-1}-1)\exp(\alpha_1 z) - \frac{g_2}{\alpha_2 Q_2}(\mu_2^{-1}-1)\exp(\alpha_2 z)\,, \quad z \leq 0 \tag{C·3a}$$

$$= -\frac{g_3}{\alpha_3 Q_3}\exp(\alpha_3 z) - \frac{g_4}{\alpha_4 Q_4}\exp(\alpha_4 z) - \frac{g_1}{\mu_1 \alpha_1 Q_1}\exp(\alpha_1 z)$$
$$- \frac{g_2}{\mu_2 \alpha_2 Q_2}\exp(\alpha_2 z)\,, \quad 0 < z \leq \sigma \tag{C·3b}$$

$$= \frac{g_3}{\alpha_3 Q_3}(\mu_3^{-1}-1)\exp(\alpha_3 z) + \frac{g_4}{\alpha_4 Q_4}(\mu_4^{-1}-1)\exp(\alpha_4 z)\,, \quad z > \sigma \tag{C·3c}$$

$$Y_s(z) = -X_s(z) + H\left(\frac{\sigma}{2} - \left|z - \frac{\sigma}{2}\right|\right) + D_X d_z^2 X_s + v d_z X_s\,, \tag{C·3d}$$

where

$$\mu_\nu = \exp(\alpha_\nu \sigma)\,.$$

Whether solutions of the above forms actually exist or not depends on whether the conditions (4·6b) and (4·7b) allow for real v and positive σ. In the extreme case as represented by Eq. (4·13) this condition is satisfied and the solution is unique in each Case (A) and (B).

Appendix D

We briefly outline the procedure of finding $\boldsymbol{u}_0^*$, together with the final expressions for $\boldsymbol{u}_0^*$, G_X and G_Y.

The quantity $\boldsymbol{u}_0^*$ is the null eigenvector of Γ^*, and the latter has in the present model the following explicit form:

$$\Gamma^* = \begin{pmatrix} -1+\delta(X_s(z)-a)+D_X d_z^2 - v d_z & b \\ -1 & -c+Dd_z^2 - v d_z \end{pmatrix}. \tag{D·1}$$

The delta function in Eq. (D·1) may explicitly be written as

$$\delta(X_s(z)-a) = -\gamma_0^{-1}\delta(z), \quad \text{(Case (A))} \tag{D·2a}$$

$$= -\gamma_\sigma^{-1}\delta(z-\sigma)+\gamma_0^{-1}\delta(z), \quad \text{(Case (B))} \tag{D·2b}$$

where

$$\gamma_{\sigma,0} = d_z X_s|_{z=\sigma,0}\,. \tag{D·3}$$

Analogously to the problem of finding X_s and Y_s, the quantity $\boldsymbol{u}_0^*$ may be expressed in terms of exponential functions $\exp(\beta_i z)$, where β_i are the zeros of the polynomial $\tilde{P}(\beta)$ defined by

$$\tilde{P}(\beta; v) = P(\beta; -v)\,. \tag{D·4}$$

Equation (D·4) implies

$$\beta_i = -\alpha_i, \; i=1, 2, 3, 4\,. \tag{D·5}$$

In terms of $x_0^*(z)$ and $y_0^*(z)$ defined by Eq. (4·10), the following conditions should now be required: The boundary conditions, $x_0^*(\pm\infty) = y_0^*(\pm\infty) = 0$; the continuity conditions for x_0^*, y_0^* and $d_z y_0^*$ at the junction points; the jump condition

$$d_z x_0^*|_{+0} - d_z x_0^*|_{-0} = (D_X\gamma_0)^{-1} x_0^*(0), \quad \text{(Case (A))} \tag{D·6}$$

and

$$d_z x_0^*|_{\sigma+0} - d_z x_0^*|_{\sigma-0} = (D_X\gamma_\sigma)^{-1} x_0^*(\sigma), \tag{D·7a}$$

$$d_z x_0^*|_{+0} - d_z x_0^*|_{-0} = -(D_X\gamma_0)^{-1} x_0^*(0)\,. \quad \text{(Case (B))} \tag{D·7b}$$

With these conditions one may determine $\boldsymbol{u}_0^*(z)$ except for a multiplicative constant. The expression for the *unnormalized* $\boldsymbol{u}_0^*$ is

(A)

$$x_0^*(z) = \frac{g_3}{Q_3}\exp(-\alpha_3 z) + \frac{g_4}{Q_4}\exp(-\alpha_4 z), \quad z\leq 0 \tag{D·8a}$$

$$= -\frac{g_1}{Q_1}\exp(-\alpha_1 z) - \frac{g_2}{Q_2}\exp(-\alpha_2 z), \quad z>0 \tag{D·8b}$$

(B)

$$x_0^*(z) = \frac{g_3}{Q_3}(\mu_3-\eta)\exp(-\alpha_3 z) + \frac{g_4}{Q_4}(\mu_4-\eta)\exp(-\alpha_4 z), \quad z\leq 0 \tag{D·9a}$$

$$=\frac{\eta g_1}{Q_1}\exp(-\alpha_1 z)+\frac{\eta g_2}{Q_2}\exp(-\alpha_2 z)+\frac{\mu_3 g_3}{Q_3}\exp(-\alpha_3 z)$$

$$+\frac{\mu_4 g_4}{Q_4}\exp(-\alpha_4 z),\quad 0<z\leq\sigma \tag{D·9b}$$

$$=\frac{g_1}{Q_1}(\eta-\mu_1)\exp(-\alpha_1 z)+\frac{g_2}{Q_2}(\eta-\mu_2)\exp(-\alpha_2 z),\quad z>\sigma \tag{D·9c}$$

where

$$\eta=-\left(\frac{\mu_3 g_3}{Q_3}+\frac{\mu_4 g_4}{Q_4}\right)\Bigg/\left(\frac{g_1}{\mu_1 Q_1}+\frac{g_2}{\mu_2 Q_2}\right). \tag{D·10}$$

As for the other notations used above, see Appendix C. The quantity $y_0{}^*$ is related to $x_0{}^*$ by

$$y_0{}^*=b^{-1}(1-D_X d_z^2+v d_z)\,x_0{}^* \tag{D·11}$$

in both cases. The quantities G_X and G_Y defined in Eq. (4·12) may now be calculated, and they turn out to have the following expressions:

(A)

$$G_X=-(\widetilde{K}_{13}+\widetilde{K}_{23}+\widetilde{K}_{14}+\widetilde{K}_{24}), \tag{D·12a}$$

$$G_Y=b(K_{13}+K_{23}+K_{14}+K_{24}), \tag{D·12b}$$

(B)

$$G_X=-\{J_{13}\widetilde{K}_{13}+J_{23}\widetilde{K}_{23}+J_{14}\widetilde{K}_{14}+J_{24}\widetilde{K}_{24}-(\mu_4-\mu_3)\widetilde{K}_{34}$$

$$-\eta(\mu_1{}^{-1}-\mu_2{}^{-2})\widetilde{K}_{12}+\frac{\sigma}{2}\left(\frac{\mu_3 g_3{}^2}{Q_3{}^2}+\frac{\mu_4 g_4{}^2}{Q_4{}^2}+\frac{\eta g_1{}^2}{\mu_1 Q_1{}^2}+\frac{\eta g_2{}^2}{\mu_2 Q_2{}^2}\right)\Bigg\}, \tag{D·13a}$$

$$G_Y=b\{J_{13}K_{13}+J_{23}K_{23}+J_{14}K_{14}+J_{24}K_{24}-(\mu_4-\mu_3)K_{34}$$

$$-\eta(\mu_1{}^{-1}-\mu_2{}^{-1})K_{12}+\frac{\sigma}{2}\left(\frac{\mu_3}{Q_3{}^2}+\frac{\mu_4}{Q_4{}^2}+\frac{\eta}{\mu_1 Q_1{}^2}+\frac{\eta}{\mu_2 Q_2{}^2}\right)\Bigg\}, \tag{D·13b}$$

where

$$K_{ij}=\frac{1}{(\alpha_i-\alpha_j)Q_i Q_j}, \tag{D·14a}$$

$$\widetilde{K}_{ij}=g_i g_j K_{ij}, \tag{D·14b}$$

$$J_{ij}=1+\eta-\mu_j-\eta\mu_i{}^{-1}. \tag{D·14c}$$

References

1) N. Rashevsky, Bull. Math Biophys. **2** (1940), 15, 65, 109.
2) A. M. Turing, Phil. Trans. Roy. Soc. London **B237** (1952), 37.

3) G. Nicolis and I. Prigogine, *Self Organization in Nonequilibrium Systems* (Wiley-Interscience, New York, 1977).
4) H. Haken, *Synergetics, An Introduction* (Springer, Berlin-Heidelberg-New York, 1978).
5) A. Gierer and H. Meinhardt, Kybernetik **12** (1972), 30.
H. Meinhardt and A. Gierer, J. Cell. Sci. **15** (1974), 321.
6) Y. Kuramoto and T. Yamada, Prog. Theor. Phys. **56** (1976), 679.
T. Yamada and Y. Kuramoto, Prog. Theor. Phys. **56** (1976), 681.
7) O. E. Rössler, Z. Naturforsch. **31a** (1976), 1168.
8) Y. Kuramoto, Prog. Theor. Phys. Suppl. No. 64 (1978), 346.
9) L. D. Landau, J. Exp. Theor. Phys. **14** (1944), 240.
10) G. I. Barenblatt, Ya. B. Zel'dovich and A. G. Istrasov, J. Appl. Mech. Tech. Phys. 4 (1962), 21.
11) G. I. Sivashinsky, Combustion Sci. Tech. **15** (1977), 137; Acta Astronautica **4** (1977), 1177.
12) G. I. Sivashinsky, SIAM J. Appl. Math. (in press).
13) J. M. Burgers, *The Nonlinear Diffusion Equation* (D. Reidel Publ. Co. Dordrech-Boston, 1974).
14) J. Rinzel and J. B. Keller, Biophys. J. **13** (1973), 1313.
15) A. T. Winfree, In *Theoretical Chemistry* **4** (Academic Press, New York - San Francisco-London, 1978), 1.
16) S. Koga and Y. Kuramoto, Prog. Theor. Phys. **63** (1980), 106.