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Pattern synthesis from singular solutions in the Debye limit: helical waves and twisted toroidal scroll structures

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Abstract. Applying the Debye limit to a suitable superposition of singular solutions of the linear two-component reaction–diffusion equations in three dimensions leads to helical and twisted toroidal scroll waves of constant concentration. An integral representation of the solutions of the Helmholtz equation in toroidal coordinates establishes a connection between helical and toroidal waves.

1. Introduction

Pattern formation in physiochemical systems through the interaction of spatial diffusion and the local properties of the chemical reactions involved has been the subject of active research in recent years (Gmitro and Scriven 1966, Nicolis and Prigogine 1977, Winfree 1980). While the role of diffusion in such a synthesis is open to debate (Thones 1973), the reaction–diffusion (R-D) hypothesis has been successful in generating a variety of testable global and local geometrical concepts in relation to possible patterns (Auchmuty and Nicolis 1976).

The object of this investigation is to show that there are several features in common between the geometrical properties of the concentration contours implied by the Debye limit (Abramowitz and Stegun 1965, p 366) of the singular solutions of the linear R-D equations and experimentally observed patterns in Belousov–Zhabotinski-type (Winfree 1980, p 300) reactions. This then suggests that the class of nonlinear models that may be relevant to observed patterns is the one that stabilises the above-mentioned concentration contours. In our opinion, linear models, despite their well-documented defects (Tyson 1976), might contain valid information on geometrical aspects of concentration contours that could be usefully incorporated into nonlinear generalisations. The emphasis here is not on amplitudes of concentrations or the facility of superposition intrinsic to linear models, but on the structure of functionals that emerge which are candidates for describing experimentally observed concentration contours.

DeSimone, Beil and Scriven (1973) (referred to hereinafter as DBS) have provided such a beginning by constructing a two-dimensional model in which the singular solutions have been taken as elementary pattern functions. They were able to obtain Archimedean spiral patterns in the asymptotic limit of large distances. Admittedly, singularity of the solution is a defect of the model. But the intriguing result is that the singular solution considered in the Debye limit leads to the well-known involute of a circle for concentration contours (see § 2). This encourages one to apply such considerations to three dimensions. The present analysis reveals that helical and twisted toroidal scroll patterns are supported by the R-D equations (2.1), and that the latter are linear functionals of the former in the Debye limit.

The contribution of this investigation is threefold:

(i) A reassessment of the DBS model in the light of the Debye limit (§ 2).

(ii) The emergence of orthonormal helical wave (Garavaglia and Gomatam 1975a,b) propagation in the Debye limit. An interesting feature of these helical waves is that orthonormal propagation is maintained only outside a singular cylinder whose radius is determined by the parameters of the model and an arbitrary separation constant (§ 3).

(iii) (a) Formulation and perturbative solution of the problem in toroidal coordinate systems. The dominant mode of propagation turns out to be a twisted toroidal scroll wave with singularities along the central axis as well as along the core axis (§ 4). An integral representation of the toroidal waves in terms of helical waves of varying pitch, phase and amplitude provides estimates on correction terms (appendix 3).
(b) An exact solution for the toroidal structure (with baffle) is briefly discussed in appendix 2.

2. Pattern synthesis from singular solutions in the Debye limit

2.1. Superposition of singular solutions

The object of this section is to establish the notation (Gmitro and Scriven 1966) and to outline the procedure for the superposition of linearly independent, singular harmonic solutions of the R-D system (DBS)

$$\partial C/\partial t = D\nabla^2 C + AC \tag{2.1a}$$

where

$$C = \begin{pmatrix} C_1 \\ C_2 \end{pmatrix} \qquad D = \begin{pmatrix} D_1 & 0 \\ 0 & D_2 \end{pmatrix} \qquad A = [a_{ij}]. \tag{2.1b}$$

The D_i and a_{ij} are real constants, to be restrained as shown below in (2.5). The C_i are to be construed as small deviations of concentrations from a possible equilibrium value of the full nonlinear R-D equations.

We look for solutions of (2.1) of the type

$$C_i = P_i^{(+)} F^{(+)}(\boldsymbol{r})\, \mathrm{e}^{-j\omega t} + P_i^{(-)} F^{(-)}(\boldsymbol{r}) \mathrm{e}^{j\omega t} \qquad (i = 1, 2) \tag{2.2}$$

where $F^{(+)}$ and $F^{(-)}$ are 'outgoing' and 'incoming' singular solutions of the Helmholtz equation

$$\nabla^2 F + k^2 F = 0 \qquad (k \text{ real}). \tag{2.3}$$

Substituting (2.2) in (2.1) and using (2.3) we obtain the eigenvector equation

$$(A - k^2 D \pm j\omega I) P^{(\pm)} = 0 \tag{2.4}$$

where $P^{(\pm)}$ is the column vector

$$\begin{pmatrix} P_1^{\pm} \\ P_2^{\pm} \end{pmatrix}.$$

The conditions

$$k^2 = \mathrm{Tr}A/\mathrm{Tr}D \tag{2.5a}$$

$$\det(A - k^2 D) > 0 \tag{2.5b}$$

guarantee that ω is real and has the value given by

$$\omega = +[\det(A - k^2 D)]^{1/2}. \tag{2.5c}$$

Since $(F^+)^* = F^-$, we must have $(P_i^+)^* = P_i^{(-)}$ in order that the C_i are real.

From (2.4) we obtain

$$\frac{P_2^{(+)}}{P_1^{(+)}} = -\frac{a_{11} - k^2 D_1 + j\omega}{a_{12}} = \left(\frac{P_2^-}{P_1^-}\right)^*. \tag{2.5d}$$

To simplify our presentation we choose

$$P_1^+ = P_1^- = \tfrac{1}{2}A_1 \tag{2.5e}$$

a real constant. Therefore

$$C_1(\mathbf{r}, t) = \tfrac{1}{2}A_1[F^{(+)}(\mathbf{r})\mathrm{e}^{-j\omega t} + F^{(-)}(\mathbf{r})\mathrm{e}^{j\omega t}]. \tag{2.6}$$

The expression for $C_2(\mathbf{r}, t)$ is obtained from (2.2) with the help of (2.5*d*, *e*).

2.2. *The Debye limit illustrated in two dimensions*

DBS have shown that a suitable combination of regular and singular solutions of (2.1*a*) in polar coordinates leads asymptotically to Archimedean spiral patterns for phases of concentrations. In this section we show that an application of the Debye limit to Bessel functions leads to the involute of a circle for the phase contours, thereby providing a precise relationship between the radius of the involute circle, the wavenumber k and the order of the Bessel functions.

In two dimensions with

$$\nabla^2 \equiv \frac{\partial^2}{\partial r^2} + \frac{1}{r}\frac{\partial}{\partial r} + \frac{1}{r^2}\frac{\partial^2}{\partial \varphi^2} \tag{2.7}$$

we obtain

$$C_1 = A_1 |H_m^{(1)}(kr)| \cos[\delta_m(kr) + m\varphi - \omega t] \qquad (m \text{ integer}) \tag{2.8}$$

where the notation (Abramowitz and Stegun 1965, p 358) is as follows:

$$|H_m^{(1)}(kr)| = [J_m^2(kr) + Y_m^2(kr)]^{1/2} \tag{2.9}$$

$$\delta_m(kr) = \tan^{-1}(Y_m/J_m). \tag{2.10}$$

In the Debye limit (Abramowitz and Stegun 1965, p 366)

$$kr,\ m \to \text{large} \qquad kr > m \tag{2.11}$$

we obtain

$$\delta_m(kr) \to m\left[\frac{[r^2 - (m/k)^2]^{1/2}}{(m/k)} - \cos^{-1}\left(\frac{m/k}{r}\right) - \frac{\pi}{4}\right] \equiv \delta_m^{\mathrm{D}}(kr) \tag{2.12}$$

where terms $O(1/m)$ have been omitted in the limit for $H_m(kr)$. The contours

$$\delta_m^D(kr)+m\varphi-\omega t=\text{constant} \tag{2.13}$$

are involute m spirals unwinding from a circle of radius

$$r_0=m/k=m\lambda/2\pi \tag{2.14}$$

where λ is the wavelength measured along the tangent

$$\varphi-\cos^{-1}\left(\frac{m/k}{r}\right)=\text{constant}. \tag{2.15}$$

It is interesting to point out that the orthonormal curvilinear system (2.13) and (2.15) is a degenerate case of the more general triply orthonormal system of helical surfaces (Garavaglia and Gomatam 1975a,b) we have considered elsewhere in a different context. We will return to this point in the next section.

An objection that has to be met is the relevance of the Debye limit (2.11) to experimentally observed single spirals, $m=1$. It is known that the Debye limit is true even for finite values of m, and this has been emphasised by Watson (1966, p 224). In fact, as an estimate (appendix 1) we have

$$\left|\delta_m(kr)-\delta_m^D(kr)\right|\sim\left|\frac{3\cot\beta+5\cot^3\beta}{24m}+O\left(\frac{1}{m^3}\right)\right|$$

where

$$kr=m\sec\beta \qquad (0<\beta<\tfrac{1}{2}\pi). \tag{2.16}$$

Numerical calculations show that the relative error associated with (2.16) when $m=1$ is less than 1% for $kr>5$ (see table A1, appendix 1).

In the next two sections we use the Debye limit to analyse the three-dimensional patterns implied by the singular solutions (2.2).

3. Linear R-D systems in three dimensions: helical patterns

It is a straightforward matter to consider any well-established coordinate system with its separable (or partially separable) singular solutions for real k and examine it in the Debye limit. One usually finds that the linear dimension exhibits a transition value, parametrised by k and the separation constants associated with other coordinates of the system. An application of these ideas to cylindrical and toroidal coordinate systems reveals many interesting features of the possible patterns. Firstly we discuss the problem in cylindrical coordinates and show how helical patterns can be constructed from singular solutions.

The system (2.1) is trivially solved when

$$\nabla^2\equiv\frac{\partial^2}{\partial r^2}+\frac{1}{r}\frac{\partial}{\partial r}+\frac{1}{r^2}\frac{\partial^2}{\partial\varphi^2}+\frac{\partial^2}{\partial z^2}. \tag{3.1}$$

The solution C_1 is

$$C_1=\tfrac{1}{2}A_1[J_m^2(qr)+Y_m^2(qr)]^{1/2}\cos[\delta_m(qr)+m\varphi-pz-\omega t] \tag{3.2a}$$

where

$$q=(k^2-p^2)^{1/2} \qquad (|p|<|k|). \tag{3.2b}$$

The choice $p = 0$ represents cylindrical scroll waves. In general we have a helical surface

$$\delta_m[(k^2 - p^2)^{1/2} r] + m\varphi - pz - \omega t = \text{constant} \tag{3.3}$$

for phase surfaces, the pitch of the helical surface being $2\pi m/p$. A wavefront is usually one of the surfaces of a triple orthonormal system of surfaces; one would certainly expect it to be so in a linear system (2.1) with constant coefficients. It is not obvious that (3.3) could be embedded in a triple orthonormal system (Eisenhart 1960) of surfaces. (The problem is a trivial one for two-dimensional curvilinear systems.) However, in the Debye limit we obtain, for a given t,

$$\delta_m^{\mathrm{D}}[(k^2 - p^2)^{1/2} r] + m\varphi - pz = \text{constant}. \tag{3.4}$$

We have shown elsewhere (Garavaglia and Gomatam 1975a, b) that the helical surface (3.4) is one of the surfaces of a triple orthonormal system of surfaces outside a singular cylinder of radius

$$r_0 = m/(k^2 - p^2)^{1/2}. \tag{3.5}$$

Thus the R–D system (2.1)–(2.3) permits orthonormal propagation of helical wavefronts in the Debye limit. The propagation vector is along the lines of curvature, the curve of intersection of tangential half-planes to the singular cylinder (3.5) and a helical surface with its sense opposite to that of (3.4). Note that z = constant sections of (3.4) are involute spirals. There is very little experimental information available on helical patterns, even though a few observed patterns in test tubes could have been interpreted either as slant bands or helical patterns. The restriction (3.2*b*) is necessary for helical waves.

Since p is arbitrary, it is natural to look into the possibility of superposing waves such as (3.2*a*). For a class of weight functions and contours such a superposition leads to toroidal scrolls with twists.

4. Toroidal scrolls with twists

The occurrence of toroidal scroll patterns has been inferred from experiments with millipore stacks soaked in Belousov–Zhabotinski (B–Z) reagents (Winfree 1973). Recently, they have been confirmed *in situ* by Burgess and Welsh (1981) in experiments with suitably modified B–Z reagents. In this section we show that a twisted scroll ring with a singularity on the hole axis emerges as a surface of constant concentration.

We tackle this problem in two stages. Firstly we consider the singular solutions of the R–D system (2.1) in a suitably constructed toroidal coordinate system. Twisted scroll ring surfaces obtain in the limit of large values of R_0, the major radius. Next we show that these could be obtained by a suitable superposition of the helical waves (3.2*a*), considered in the Debye limit. From this, we obtain estimates on corrections to the dominant twisted scroll ring pattern. The method also elucidates the nature of the singularity located along the hole axis.

While the present investigation was well under way, we came across a wealth of literature on electromagnetic wave propagation (Lewin 1974, Lewin *et al* 1977) in toroidal structures, the primary concern of these investigations being to study the phenomenon of radiation losses due to curvature in the structure. such as a curved dielectric fibre. When dealing with such problems a number of well-tested physical considerations and boundary conditions on the surfaces of waveguides dictate the choice of solutions. In our case the chief aim is to investigate the Debye-asymptotic geometry of wave patterns concealed in the singular solutions of the R-D equations in toroidal coordinates. However, the methods outlined in the references stated above could be applied to our problem with suitable modifications.

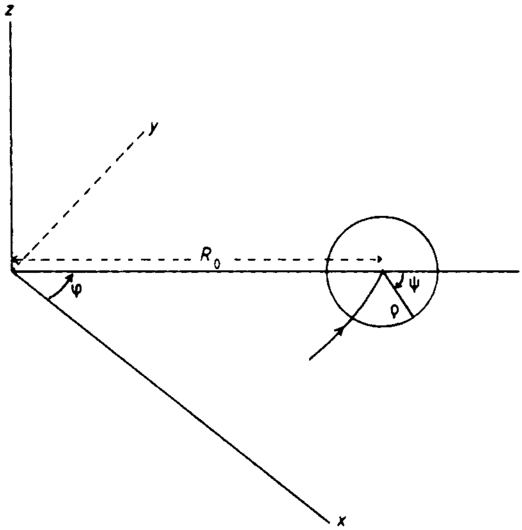


Figure 1. Toroidal coordinate system.

The toroidal coordinate system (Lewin *et al* 1977) is shown in figure 1. The system (ρ, ψ, φ) is related to the cylindrical coordinates (r, z, φ) by

$$r = R_0 + \rho \cos \psi \tag{4.1a}$$

$$z = -\rho \sin \psi. \tag{4.1b}$$

The half-planes φ = constant are common to both systems. It is easily seen that the toroidal surfaces $\rho = c_1$ intersect the half-planes φ = constant in a circle whose centre is at $r = R_0$. As we vary c_1 we obtain a series of concentric circles for $c_1 < R_0$ and segments of concentric circles for $c_1 > R_0$. This coordinate system should be contrasted with the classical toroidal systems (Lebedev 1965) which include a system of eccentric circles.

The Helmholtz equation (2.3) takes the following form in the toroidal system:

$$\left[\frac{\partial^2}{\partial\rho^2} + \frac{2 - R_0/r}{\rho}\frac{\partial}{\partial\rho} + \frac{1}{\rho^2}\frac{\partial^2}{\partial\psi^2} - \left(1 - \frac{R_0}{r}\right)\frac{\tan\psi}{\rho^2}\frac{\partial}{\partial\psi} + \frac{1}{r^2}\frac{\partial^2}{\partial\varphi^2} + k^2\right]F = 0 \tag{4.2}$$

where we recall that $r = R_0 + \rho \cos \psi$. An immediate separation of $G(\rho, \psi)$ in the φ coordinate is possible if we make the assumption

$$F = G(\rho, \psi)\, e^{i\nu\varphi} \qquad (\nu \neq 0) \tag{4.3}$$

$$\left[\frac{\partial^2}{\partial\rho^2} + \frac{2 - R_0/r}{\rho}\frac{\partial}{\partial\rho} + \frac{1}{\rho^2}\frac{\partial^2}{\partial\psi^2} - \left(1 - \frac{R_0}{r}\right)\frac{\tan\psi}{\rho^2}\frac{\partial}{\partial\psi} + k^2 - \frac{\nu^2}{r^2}\right]G = 0. \tag{4.4}$$

The presence of the factor $1/r$ does not allow any further separation of $G(\rho, \psi)$. (However, when $\nu = \frac{1}{2}$, the system (4.4) has the solutions

$$G = \frac{1}{r^{1/2}} H_l^{(1,2)}(k\rho)\, e^{\pm i\psi l}.$$

These solutions, relevant to a toroidal structure with a baffle located at $\varphi = \varphi_1$, are derived in appendix 2.) One possible approach is to use perturbation methods applied to the parameter ν/R_0. But a more instructive way is to construct integral representations for solutions of (4.4), using the known exact solutions in cylindrical coordinates.

Assuming that $R_0 \gg |\rho \cos\psi|$, we expand $1/r^2$ to give

$$\frac{1}{r^2} \approx \frac{1}{R_0^2} - \frac{2\rho\cos\psi}{R_0^3}. \tag{4.5}$$

Then (4.4) reduces to

$$\left[\frac{\partial^2}{\partial\rho^2} + \frac{1}{\rho}\frac{\partial}{\partial\rho} + \frac{1}{\rho^2}\frac{\partial^2}{\partial\psi^2} + k^2 - \frac{\nu^2}{R_0^2} + \frac{1}{R_0}\left(\cos\psi\frac{\partial}{\partial\rho} - \frac{1}{\rho}\sin\psi\frac{\partial}{\partial\psi} + \frac{2\nu^2}{R_0^2}\rho\cos\psi\right)\right]G_A = 0. \tag{4.6}$$

Here G_A represents the solutions under assumption (4.5). We are interested in propagating solutions in the local coordinate system (ρ, ψ); hence we stipulate that

$$k^2 - \nu^2/R_0^2 > 0. \tag{4.7}$$

A further assumption

$$kR_0 > 1 \tag{4.8}$$

enables us to neglect the terms containing $1/R_0$ in (4.6). In the local coordinate system (ρ, ψ) we have

$$\left(\frac{\partial^2}{\partial\rho^2} + \frac{1}{\rho}\frac{\partial}{\partial\rho} + \frac{1}{\rho^2}\frac{\partial^2}{\partial\psi^2} + k_0^2\right)G^{((0))} = 0 \tag{4.9}$$

where

$$k_0 = (k^2 - \nu^2/R_0^2)^{1/2}. \tag{4.10}$$

Hence the solution (4.3) now is approximated by

$$F_{\nu,l}^{((0))} = H_l^{(1,2)}(k_0\rho)\, e^{\pm il\psi}\, e^{\pm i\nu\varphi}. \tag{4.11}$$

The phase contours (2.2) are now determined by

$$C_1 = P_1^{(+)} H_l^{(1)}(k_0\rho)\, e^{i(\nu\varphi + l\psi - \omega t)} + P_1^{(-)} H_l^{(2)}(k_0\rho)\, e^{-i(\nu\varphi + l\psi - \omega t)} \tag{4.12}$$

$$= A_1 |H_l^{(1)}(k_0\rho)| \cos[\delta_l(k_0\rho) + \nu\varphi + l\psi - \omega t]. \tag{4.13}$$

A consideration of the Debye limit

$$k_0\rho,\ l \to \text{large} \qquad k_0\rho > l$$

with the implied restriction $|\rho\cos\psi| \ll R_0$ results in the constant phase surfaces

$$l\left[\frac{[\rho^2 - (l/k_0)^2]^{1/2}}{l/k_0} - \cos^{-1}\left(\frac{l/k_0}{\rho}\right)\right] + l\psi + \nu\varphi - \omega t = \text{constant}. \tag{4.14}$$

Consider the surface (4.14) at a particular instant of time $t = \bar{t}$.

Case $l=1$, $\nu=0$. The equation (4.14) represents a scroll surface, the scroll being wound on a torus of major radius R_0 and minor radius $1/k_0$ (core radius). The intersection of the half-plane $\varphi=\bar{\varphi}$ and the scroll surface is an involute 1-spiral $\delta_1^{\mathrm{D}}(k_0\rho)+\psi=\text{constant}+\omega\bar{t}$. This spiral emerges from the point $(\rho, \psi, \varphi)\equiv(1/k_0, \bar{\psi}, \bar{\varphi})$ where $\bar{\psi}=\text{constant}+\omega\bar{t}$. The involute spiral on the plane $\varphi=\bar{\varphi}+\pi$ is the mirror image of the spiral on the plane $\varphi=\bar{\varphi}$, the mirror coinciding with the planes $\varphi=\bar{\varphi}+\frac{1}{2}\pi$ and $\varphi=\bar{\varphi}+\frac{3}{2}\pi$. This double spiral concentration contour has been observed in a number of experiments (Winfree 1980, p 313).

Case $l=1$, $\nu=1$. The intersection of the half-plane $\varphi=\bar{\varphi}$ and (4.14) is again an involute 1-spiral $\delta_1^{\mathrm{D}}(k_0\rho)+\psi=\text{constant}+\omega\bar{t}-\bar{\varphi}$, but now emerging from the point $(1/k_0, \bar{\psi}, \bar{\varphi})$ on the circumference of the core where $\bar{\psi}=\text{constant}+\omega\bar{t}-\bar{\varphi}$. But the intersection of the half-plane $\varphi=\bar{\varphi}+\pi$ with the surface (4.14) is an involute 1-spiral emerging from $(1/k_0, \bar{\psi}-\pi, \bar{\varphi}+\pi)$. Thus the locus of points of emergence of the involute spirals is a helix drawn on the surface of the torus. The once-twisted toroidal scroll surface could be visualised as follows: Imagine a string wound around a torus of minor radius $1/k_0$ in a helix, so that the string retraces the helical path several times. Now unwind the string by holding one end, making sure that it is kept taut all the time. The surface generated by the unwinding string is a toroidal scroll with one twist! Twisted toroidal scroll surfaces have been suggested as possible candidates for concentration contours (Winfree 1980, pp 254–6).

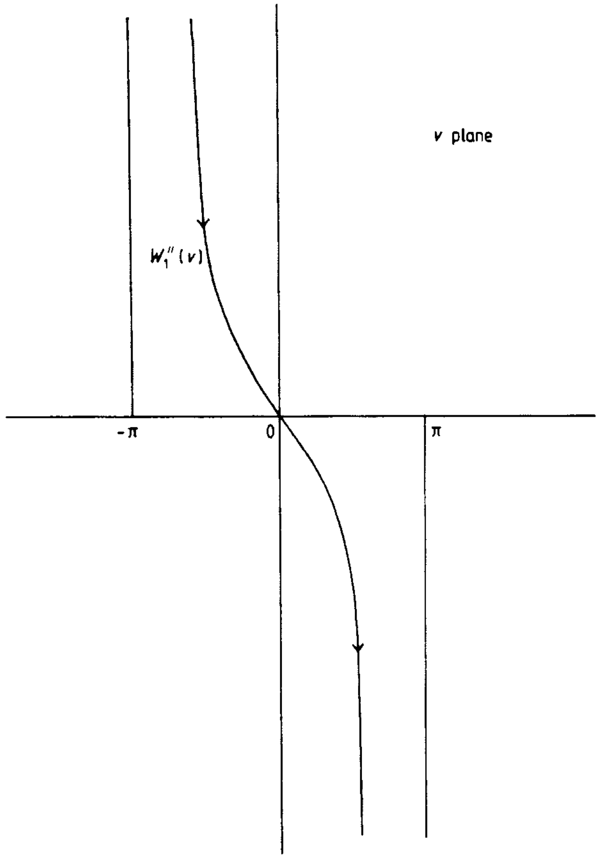
The singularity at $r=0$ is not evident in (4.13). This is not surprising since (4.12) is a local description of the solution, with $|\rho\cos\psi|\ll R_0$. An answer to this question emerges when we consider the method of integral representation (Lewin *et al* 1977) of the solution. We start with a superposition of solutions in cylindrical coordinates

$$\mathrm{e}^{\mathrm{i}\nu\varphi}\int_{W_1(p)} H_\nu^{(1)}[(k^2-p^2)^{1/2}r]\,\mathrm{e}^{-\mathrm{i}pz}\,Q_\nu^{(+)}(p)\,\mathrm{d}p. \tag{4.15}$$

The problem now is to choose the weight function $Q_\nu^+(p)$ and the contour $W_1(p)$ suitably to produce the solution (4.11) in the local coordinate system. Of course $\mathrm{e}^{\pm\mathrm{i}\nu\varphi}$ drops out of this consideration. We first consider the Hankel function $H_\nu^{(1)}[(k^2-p^2)^{1/2}r]$ in the Debye limit, then seek the Taylor expansion of the Debye-asymptotic form around $r=R_0$ for small values of $|\rho\cos\psi|$. It turns out that terms of order $\rho\cos\psi$ in the exponent are enough to yield the solution (4.11) when $W_1(p)$ and $Q_\nu^+(p)$ are suitably chosen. The correction terms are of the order $(\rho\cos\psi)/R_0$. The details of the calculations and the concepts involved are sufficiently different from electromagnetic problems to be presented here in separate appendices. The main result is (see appendix 3)

$$\begin{aligned}\frac{1}{\pi}\int_{W_1'(u)}\mathrm{d}u\bigg(P_1^{(+)}&\frac{H_\nu^{(1)}[(k_0\cos u)r]}{H_\nu^{(1)}[(k_0\cos u)R_0]}\exp\{\mathrm{i}[l(u-\tfrac{1}{2}\pi)+\nu\varphi-k_0z\sin u-\omega t]\}\\ &-P_1^{(-)}\frac{H_\nu^{(2)}[(k_0\cos u)r]}{H_\nu^{(2)}[(k_0\cos u)R_0]}\exp\{-\mathrm{i}[l(u-\tfrac{1}{2}\pi)+\nu\varphi-k_0z\sin u-\omega t]\}\bigg)\\ &\approx P_1^{(+)}H_l^{(1)}(k_0\rho)\,\mathrm{e}^{\mathrm{i}(\nu\varphi+l\psi)}+P_1^{(-)}H_l^{(2)}(k_0\rho)\,\mathrm{e}^{-\mathrm{i}(\nu\varphi+l\psi)}\end{aligned} \tag{4.16}$$

for $|\rho\cos\psi|\ll r$ where correction terms of the order $(\rho\cos\psi)/R_0$ are neglected in the integral as shown in appendix 3. The contour $W_1'(u)$ is related to the contour $W_1''(v)$ (Sommerfeld 1949) for integral representation of $H_l^{(1)}(\eta)$ (see figure 2) by the



17051 **Figure 2.** The contour $W_1''(v)$ for integral representation.

transformation $u = v + i\psi$ as shown in appendix 3. The integral is easily recognised to be a superposition of helical waves of varying pitches, phases and amplitudes. The singularity at $r = 0$ enters through $H_\nu^{(1,2)}[(k^2 - p^2)^{1/2} r]$.

The case $\nu = 0$ is discussed in appendix 3. Since there is no transition radius, a straightforward asymptotic limit of $H_0^{(1,2)}[(k^2 - p^2)^{1/2} r]$ generates the right solution in the local coordinate system.

5. Discussion

The role of singular solutions had been well understood in the context of classical boundary value problems (Sommerfeld 1949). Various perturbative procedures, such as eikonal methods, have provided important physical insights into the effect of potential barriers or material boundaries on the solution. The Debye limit is an offshoot of such procedures. In the absence of such boundaries or given the intriguing behaviour of B–Z wavefronts at such boundaries (Winfree 1980, p 300), the Debye limit gives quantitative information on the self-organisation of the system into at least two domains with distinct properties.

Perhaps it is this information on phase curves that is the unifying feature of linear and nonlinear models. The λ–ω type systems (Kopell and Howard 1981) in higher dimensions would provide a natural framework for incorporating this unifying feature.

The emergence of helical wave patterns raises questions which have experimental implications. How does one produce and maintain these patterns, and by what methods does one observe and characterise various planar projections of these helical waves? An equally intriguing question is why in many observations to date the toroidal scroll is favoured by the chemical system. It is important to answer these questions, especially in view of the connection we have established between the helical waves and twisted toroidal scroll structures.

An insight into the pattern formation resulting in extended structures like toroidal scrolls might be gained from applying bifurcation (Nicolis and Prigogine 1977, p 178) techniques to the exact solutions we have outlined in appendix 2. The ultimate objective of this analysis would be to relate the major radius R_0 of the toroidal structure to the internal parameters of the system. These ideas are currently under investigation.

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Appendix 1. The Debye limit (Abramowitz and Stegun 1965 p 366, Watson 1966, p 244)

The Hankel function in the Debye limit $kr > \nu$, kr and ν large, is given by

$$H_\nu^{(1)}(\nu \sec\beta) = \left(\frac{2(L^2+M^2)}{\pi\nu \tan\beta}\right)^{1/2} \exp\left[i\left(\Psi - \tan^{-1}\frac{M}{L}\right)\right]$$

where

$$kr = \nu \sec\beta \qquad (0 < \beta < \tfrac{1}{2}\pi \quad \nu > 0)$$

$$\Psi = \nu(\tan\beta - \beta) - \tfrac{1}{4}\pi$$

$$L \sim 1 - \frac{81\cot^2\beta + 462\cot^4\beta + 385\cot^6\beta}{1152\nu^2} + O\left(\frac{1}{\nu^4}\right)$$

$$M \sim \frac{3\cot\beta + 5\cot^3\beta}{24\nu} + O\left(\frac{1}{\nu^3}\right).$$

For ν large

$$\tan^{-1}\frac{M}{L}\sim\tan^{-1}\left\{\left[\frac{3\cot\beta+5\cot^3\beta}{24\nu}+O\left(\frac{1}{\nu^3}\right)\right]\right.$$
$$\left.\times\left[1+\frac{81\cot^2\beta+462\cot^4\beta+385\cot^6\beta}{1152\nu^2}+O\left(\frac{1}{\nu^4}\right)\right]\right\}$$
$$\sim\frac{3\cot\beta+5\cot^3\beta}{24\nu}\left(1+\frac{81\cot^2\beta+462\cot^4\beta+385\cot^6\beta}{1152\nu^2}\right)$$

where the argument of $\tan^{-1}\{\}$ is small. Hence

$$\tan^{-1}\frac{M}{L}\sim\frac{3\cot\beta+5\cot^3\beta}{24\nu}+O\left(\frac{1}{\nu^3}\right).$$

Watson (1966, pp 224, 513, 514) emphasises that many of the results derived for large ν happen to be true for functions of all positive orders. Numerical calculations indicate that for values as low as $\nu = 1, 2$ the relative error

$$(\mathrm{RE})_\nu^{\mathrm{D}}=\frac{|\delta_\nu(kr)-\delta_\nu^{\mathrm{D}}(kr)|}{\delta_\nu(kr)}\times 100$$

is less than 1% in the domain $kr_\nu < kr < \infty$ (see table A1).

Table A1. Comparison between $\delta_\nu(kr)$ and $\delta_\nu^{\mathrm{D}}(kr)$ for $\nu = 1, 2$.

ν	kr	$\delta_\nu(kr)$	$\delta_\nu^{\mathrm{D}}(kr)$
1	5.000	2.718	2.744
	6.000	3.706	3.727
	8.000	5.690	5.706
	10.000	7.681	7.694
2	6.200	2.574	2.598
	7.200	3.532	3.553
	8.200	4.501	4.518
	9.200	5.476	5.491

Appendix 2. Exact solution for toroidal coordinates ($\nu = \pm\frac{1}{2}$)

The transformation

$$G(\rho, \psi)=\frac{(r-R_0)^\mu}{r^{1/2}}U(\rho, \psi)$$

reduces (4.3) to

$$\frac{\partial^2 U}{\partial\rho^2}+\frac{2\mu+1}{\rho}\frac{\partial U}{\partial\rho}-\frac{2\mu\tan\psi}{\rho^2}\frac{\partial U}{\partial\psi}+\frac{1}{\rho^2}\frac{\partial^2 U}{\partial\psi^2}+\left(\frac{\mu(\mu-1)}{(r-R_0)^2}+\frac{\frac{1}{4}-\nu^2}{r^2}+k^2\right)U=0.$$

Choosing $\mu=0$ and $\nu=\pm\frac{1}{2}$, we obtain the Bessel equation

$$\frac{\partial^2 U}{\partial\rho^2}+\frac{1}{\rho}\frac{\partial U}{\partial\rho}+\frac{1}{\rho^2}\frac{\partial^2 U}{\partial\psi^2}+k^2U=0.$$

Appendix 3. The derivation of equation (4.16)

Consider the Debye expansion (Abramowitz and Stegun 1965, p 366) of $H_\nu^{(1)}(\eta)$ for $|\eta|>\nu$:

$$H_\nu^{(1)}(\eta)\sim \mathrm{e}^{g_\nu(\eta)} \tag{A3.1}$$

where

$$g_\nu(\eta)=\frac{1}{2}\ln\left(\frac{2}{\pi\nu}\right)-\tfrac{1}{2}\ln\tan\beta+\mathrm{i}[\nu(\tan\beta-\beta)-\tfrac{1}{4}\pi]$$
$$+\ln\left[1-\mathrm{i}\frac{3\cot\beta+5\cot^3\beta}{24\nu}+\mathrm{O}\left(\frac{1}{\nu^2}\right)\right] \tag{A3.2}$$

and

$$\eta=\nu\sec\beta \qquad (0<\beta<\tfrac{1}{2}\pi).$$

The Taylor expansion of $g_\nu(\eta)$ about η for small ε leads to

$$H_\nu^{(1)}(\eta+\varepsilon)\sim H_\nu^{(1)}(\eta)\,\mathrm{e}^{\mathrm{i}\varepsilon\,\nu(\tan\beta)/\eta}$$
$$\times\left[1-\frac{\varepsilon}{2\eta}\left(\frac{\sec\beta}{\tan\beta}\right)^2+\frac{3\varepsilon\mathrm{i}}{\eta}\frac{\operatorname{cosec}^2\beta(\cot\beta+5\cot^3\beta)}{24\nu-\mathrm{i}(3\cot\beta+5\cot^3\beta)}+\mathrm{O}\left(\frac{1}{\nu^2}\right)\right]. \tag{A3.3}$$

Setting $\eta=qR_0$, $\varepsilon=q\rho\cos\psi$ we obtain

$$H_\nu^{(1)}(qR_0+q\rho\cos\psi)\sim H_\nu^{(1)}(qR_0)\exp[\mathrm{i}\rho\cos\psi(q^2-\nu^2/R_0^2)^{1/2}]$$
$$\times\left[1-\frac{\varepsilon\eta^2}{2R_0(\eta^2-\nu^2)}+\varepsilon\mathrm{O}\left(\frac{1}{\nu}\right)\right]. \tag{A3.4}$$

Therefore, to order $\rho\cos\psi$, the dominant contribution comes from the first term on the right-hand side of (A3.4).

Thus

$$F_\nu^{(+)}=\int_{W_1(p)}\mathrm{d}p\,Q_\nu^{(+)}(p)H_\nu^{(1)}(qR_0)\exp\left[\mathrm{i}\rho\cos\psi\left(q^2-\frac{\nu^2}{R_0^2}\right)^{1/2}\right]\mathrm{e}^{-\mathrm{i}pz}\,\mathrm{e}^{\mathrm{i}\nu\varphi}. \tag{A3.5}$$

With

$$p=k_0\sin u \qquad q=(k_0^2-k_0^2\sin^2 u)^{1/2}\equiv k_0\cos u$$

we obtain

$$F_\nu^{(+)}=\mathrm{e}^{\mathrm{i}\nu\varphi}\int_{W_1'(u)}\mathrm{d}u\,k_0\cos u\,Q_\nu^{(+)}(p)H_\nu^{(1)}(qR_0)\,\mathrm{e}^{\mathrm{i}k_0\rho\cos(u-\psi)}.$$

For a general $Q_\nu^{(+)}(p)$ there is a cut in the ν plane at $u=\pm\frac{1}{2}\pi$ corresponding to $q=0$. Rather than taking the reader through tedious transformations on an initial contour, we will display the final orientation of the contour (figure 2). The cuts at $u=\pm\frac{1}{2}\pi$ will pose no problem as they will disappear when an appropriate weight function $Q_\nu^{(+)}$ is chosen.

Let

$$Q_\nu^{(+)}(p)H_\nu^{(1)}(qR_0)k_0\cos u=\pi^{-1}\,e^{il(u-\frac{1}{2}\pi)}.$$

Then

$$\begin{aligned}F^{(+)}&=\frac{e^{i\nu\varphi}}{\pi}\int_{W_1'(u)}du\;e^{ik_0\rho\cos(u-\psi)+il(u-\frac{1}{2}\pi)}\\&=\frac{1}{\pi}e^{il\psi+i\nu\varphi}\int_{W_1''(v)}dv\;e^{ik_0\rho\cos v+il(v-\frac{1}{2}\pi)}\\&=e^{il\psi+i\nu\varphi}H_l^{(1)}(k_0\rho).\end{aligned}$$

The choice $Q_\nu^{(-)}(p)H_\nu^{(2)}(qR_0)k_0\cos u=-\pi^{-1}\,e^{-il(u-\frac{1}{2}\pi)}$ and the same $W_1'(u)$ or equivalently $W_1''(v)$ leads to

$$F_\nu^{(-)}=e^{-il\psi-i\nu\varphi}H_l^{(2)}(k_0\rho).$$

The case $\nu=0$ (toroidal scroll surfaces) is dealt with by considering, for large η,

$$H_0^{(1)}(\eta)\to\left(\frac{2}{\pi\eta}\right)^{1/2}e^{i(\eta-\frac{1}{4}\pi)}\left[1-\frac{i}{8\eta}+O\left(\frac{1}{\eta^2}\right)\right]$$

and

$$H_0^{(1)}(\eta+\varepsilon)\to H_0^{(1)}(\eta)\exp\left[\varepsilon\left(i-\frac{1}{2\eta}+\frac{i/8\eta^2+O(1/\eta^3)}{1-i/8\eta+O(1/\eta^2)}\right)\right].$$

Setting $\eta=(k^2-p^2)^{1/2}R_0\equiv qR_0$, $\varepsilon=q\rho\cos\psi$ in the integral representation we determine the weight functions $Q_0^{(+)}(p)$ to be

$$Q_0^{(+)}(p)=\frac{e^{il(u-\frac{1}{2}\pi)}}{k\pi H_0^{(1)}(qR_0)\cos u}$$

where as before $p=k\sin u$ and the contour is $W_1'(u)$. A suitable combination of singular solutions would then lead to constant-concentration toroidal l scrolls

$$l\left[\frac{[\rho^2-(l/k)^2]^{1/2}}{l/k}-\cos^{-1}\left(\frac{l/k}{\rho}\right)\right]+l\psi-\omega t=\text{constant}$$

in the Debye limit. The restriction $|\rho\cos\psi|\ll R_0$ is still in force.

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