We explore the application of a pseudo-spectral Fourier method to a set of reaction-diffusion equations and compare it with a second-order finite difference method. The prototype cubic autocatalytic reaction-diffusion model as discussed by Gray and Scott [Chem. Eng. Sci. 42, 307 (1987)] with a nonequilibrium constraint is adopted. In a spatial resolution study we find that the phase speeds of one-dimensional finite amplitude waves converge more rapidly for the spectral method than for the finite difference method. Furthermore, in two dimensions the symmetry preserving properties of the spectral method are shown to be superior to those of the finite difference method. In studies of plane/axisymmetric nonlinear waves a symmetry breaking linear instability is shown to occur and is one possible route for the formation of patterns from infinitesimal perturbations to finite amplitude waves in this set of reaction-diffusion equations. © 1996 American Institute of Physics. [S1054-1500(96)00601-9]

I. INTRODUCTION

The relatively recent experimental discovery of chemical systems that undergo Turing instabilities1–7 has spurred theoretical interest in related reaction-diffusion equations and in systems where pattern formation occurs in general.5–9 Computational simulations have become a standard means of exploring the dynamics of and pattern formation that occur in equations that describe these systems.5,10–14 These simulations are often done in doubly-periodic or other simple domains. This is due to an intrinsic length scale that depends not on the size of the domain but on the dynamics of the reaction-diffusion system. This scale arises from the interaction of the nonlinear terms and nonequilibrium constraints with dissipative terms characterized by small constant diffusion coefficients. In many systems the patterns form only after finite amplitude perturbations have been introduced.

Because the phenomena we wish to explore are adequately described in a double-periodic domain, one candidate for a numerical method to solve the equations is based on an expansion in a Fourier series. In addition, the diffusion coefficient is usually assumed to be constant and thus an integrating factor may be introduced to facilitate the integration of the diffusive term implicitly with great numerical accuracy. The nonlinear term often occurs in the form of a low order polynomial in the concentrations of the chemical species and thus de-aliasing may be performed using a straightforward padding procedure. Although de-aliasing may not be as important in this system as in fluid dynamical problems with convection—because the nonlinear term contains no derivative—all computations presented here are done using a fully de-aliased method.

Here we consider a set of prototype cubic autocatalytic reaction-diffusion equations with a nonequilibrium constraint.13,9 In a spatial resolution study, we find that nonlinear wave speeds converge at a much greater rate for the pseudo-spectral method than in a calculation using a finite difference method. This is an expected result but is still an interesting illustration of the rapid convergence of spectral methods over that of finite difference methods. Although the wave speed determined with a one-dimensional finite difference method may be accurate enough for a particular application, a lower resolution spectral method might be used in its place and thus compete in terms of computational cost, even when only moderate accuracy is required.

The excellent symmetry preserving properties of spectral methods over a simple two-dimensional finite difference discretization of the Laplacian operator is well known. When an axisymmetric Gaussian disturbance is added to a spatially uniform state, the solution from the finite difference method evolves into a traveling wave with a tendency to form a square shape which eventually breaks up and forms patterns. The resolved pseudo-spectral calculation preserves the symmetry of the initial disturbance and waves remain axisymmetric until they reach a critical radius or the edge of a periodic box.

The instability of traveling waves to infinitesimal disturbances may be another mechanism for the formation of patterns not presently addressed. We find that axisymmetric traveling waves are unstable to very small perturbations. This occurs through a linear symmetry breaking instability and it may be possible to describe the phenomena in the context of a linear instability theory. However, the wave speed and amplitude of these circular traveling waves depends on the radius and thus we do not have a steady state in a strict sense about which to linearize. A linear instability theory is easily formulated for nonlinear plane waves. Numerical integration of the equations of motion displays this linear growth and the existence of a finite most unstable wavelength. This instability manifests itself when tailored or random noise is added to the initial disturbance or the traveling wave and ends when finite amplitude patterns emerge.

In Sec. II we introduce the reaction-diffusion system that
we consider, some of its properties and the standard finite difference formulation for the numerics. A thorough explanation of the pseudo-spectral Fourier method is given in Sec. III. Next, in Sec. IV, a comparison of the numerical properties of the two methods, wave speeds in one dimension and symmetry breaking properties in two dimensions is presented. Finally, Sec. V, for this system a qualitatively new phenomena has been discovered; symmetry breaking linear instabilities of one-dimensional nonlinear plane and axisymmetric waves are introduced.

II. REACTION-DIFFUSION EQUATIONS

In this paper we consider a modified form of the prototype cubic autocatalytic reaction-diffusion system as formulated by Gray and Scott.\cite{15-18} They introduce the following steps as being among the simplest representations of nonlinear chemical feedback in an isothermal system,

\begin{align}
A + 2B &\rightarrow 3B \quad \text{rate } = k_1 a b^2, \\
B &\rightarrow C \quad \text{rate } = k_2 b. 
\end{align}

(1)

(2)

The underlying stoichiometry is $A \rightarrow B$ but the reaction rate depends quadratically on the concentration of the product species, $B$. The first rate-law arises in both solution-phase and gas-phase kinetics and in biological situations. A first-order decay to a stable product, Eq. (2), sometimes occurs when the autocatalyst reacts further and introduces richer behavior.

The reaction-diffusion equations that we consider are identical to those considered by Pearson.\cite{13} The inclusion of a nonequilibrium constraint is represented by a feed term for $A$, where the feed process removes both $A$ and $B$. The concentrations are scaled by the concentration of $A$ at the inlet, time is scaled by the inverse of the reaction rate $\kappa$ of Eq. (1). The reaction-diffusion equations in nondimensional form that we will consider for the rest of the paper are:

\begin{align}
\frac{\partial a}{\partial t} &= D_A \nabla^2 a - a b^2 + F(1 - a), \\
\frac{\partial b}{\partial t} &= D_B \nabla^2 b + a b^2 - (F + \kappa) b, 
\end{align}

(3)

where $\nabla^2$ is the Laplace operator, $\kappa = k_2/k_1$ is the nondimensional rate constant of Eq. (2) and $F$ is the nondimensional feed rate. The nondimensional diffusion coefficients considered are the same as those considered by Pearson,\cite{13} $D_A = 2 \times 10^{-5}$ and $D_B = 10^{-5}$. The boundary conditions are periodic and the system size, $L$, is of order one or greater such that $L$ is greater than the intrinsic length of structures that form from finite amplitude perturbations.

When the concentrations are spatially uniform, the diffusion term may be dropped and the resulting ordinary differential equations describe the reaction kinetics. Although a detailed discussion is not appropriate here, a rich set of behavior exists for these and related ordinary differential equations. Gray and Scott\cite{16-18} include analysis of these prototype reactions while general discussions can be found in Guckenheimer and Holmes.\cite{19} Three spatially uniform steady states exist as solutions of the above equations:

\begin{align}
a &= 0, \quad \frac{1}{2(1 + \kappa/F)} \left(1 \pm \sqrt{1 - 4F(1 + \kappa/F)^2}\right), \\
b &= 1, \quad \frac{1}{2(1 + \sqrt{1 - 4F(1 + \kappa/F)^2})}.
\end{align}

(4)

(5)

The trivial steady state described by $a = 1, b = 0$ is linearly stable for all positive $F$ and $\kappa$. For regions in parameter-space where $4F(1 + \kappa/F)^2 < 1$ all three states that exist are physically relevant. In most of this region there are two stable steady states, the trivial solution and one of the nontrivial solutions. In part of this region the nontrivial stable solution loses stability and corresponds to a periodic orbit.

The full partial differential equations (3) as well as similar reaction-diffusion equations where pattern formation occurs are routinely studied using finite difference techniques.\cite{10,12,13} For our second-order finite difference method, we discretize the diffusion operator using second-order central differencing, while time is discretized using a third-order Adams-Bashforth method. Although this time-stepping scheme may not be the best choice for the diffusion term because of the restrictions on the time step for high resolutions due to stability constraints, it is simple to implement and is shown to have accuracy and stability restrictions appropriate for the nonlinear terms and can be used in both the finite difference and spectral method. This choice will not affect our comparison of the methods because the time step is chosen to be small enough to satisfy stability restrictions and correspond to errors smaller than those incurred due to a finite difference spatial discretization or a moderately resolved pseudo-spectral method.

III. PSEUDO-SPECTRAL FOURIER METHOD

Here we describe the pseudo-spectral Fourier method used to solve the reaction-diffusion equations described above. For a general discussion of spectral methods see Canuto et al.\cite{20} A number of factors exist which suggest the use of pseudo-spectral Fourier methods. The most important is the existence of an intrinsic length scale chosen by the dynamics of the system and independent of the size of the domain. By choosing an appropriately large box, we may impose periodic boundary conditions for which the natural expansion basis is a Fourier basis. Furthermore, because the diffusion coefficient is constant in space the Laplacian is easily inverted, and the analytical solution of the heat equation for the Fourier coefficients of the variables may be used in the numerical formulation. All linear terms and the spatially-temporally constant forcing terms may also be included in the analytic solution leaving only the nonlinear term to be integrated in time numerically. The nonlinear term, which is computed in physical space then transformed back to Fourier space using a discrete Fourier transform, DFT, will also introduce aliasing into the calculations if it is

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The occurrence and removal of aliasing errors is discussed in the Appendix. 

In this section we present a one-dimensional for backward differences are handled in a straightforward manner by letting the wavenumber and coordinate be orthogonal to the set of test functions. 

The concentrations, \( a \) and \( b \) from Eqs. (3), are 

\[
a(x,t) = \sum_{n=-N/2+1}^{N/2-1} \hat{a}_n(t) e^{ik_n x},
\]

where \( N - 1 \) is the total number of expansion coefficients, \( n \) is an integer, \( k_n = \alpha n \) is the wavenumber, \( \alpha = 2 \pi / L \) is the minimum nontrivial wavenumber, and \( L \) is the length of the periodic box. 

A set of ordinary differential equations for \( \hat{a}_n(t) \) and \( \hat{b}_n(t) \) are obtained by requiring that the residual of Eqs. (3) be orthogonal to the set of test functions, 

\[
S_n = \langle \epsilon^{-ik_n x} \rangle - N/2 + 1 \leq n \leq N/2 - 1.
\]

The equations of motion for each of the Fourier coefficients are 

\[
\left( \frac{d}{dt} + D_A k_n^2 + F \right) \hat{a}_n - \delta_{n,0} F = -(ab^2)_n,
\]

\[
\left( \frac{d}{dt} + D_B k_n^2 + F + \kappa \right) \hat{b}_n = (ab^2)_n,
\]

where \( \delta_{n,0} \) is the Kronecker delta and \( (ab^2)_n \) is the coefficient from the truncated Fourier expansion of the nonlinear term. We find this term by Fourier transforming \( \hat{a}_n \) and \( \hat{b}_n \) from Fourier space to physical space and doing the multiplication in physical space. We then transform back from Fourier space to integrate Eqs. (7) in time with the Fourier coefficients of the nonlinear term as a source term. In evaluating the nonlinear term we use a DFT with \( M = 2N \) coefficients to ensure that aliasing errors are removed.

Because the diffusion coefficients are constant and the nonequilibrium constraint is simple, we define an integrating factor for each of the above equations: 

\[
\hat{\chi}_n = (\hat{a}_n - \delta_{n,0}) e^{\gamma_n t}, \quad \hat{\xi}_n = \hat{b}_n e^{\lambda_n t}
\]

where 

\[
\gamma_n = D_A k_n^2 + F, \quad \lambda_n = D_B k_n^2 + F + \kappa.
\]

The equations of motion for these factors reduce to, 

\[
\frac{\partial \hat{\chi}_n}{\partial t} = -(ab^2)_n e^{\gamma_n t}, \quad \frac{\partial \hat{\xi}_n}{\partial t} = (ab^2)_n e^{\lambda_n t}.
\]

It is these equations that are discretized in time. 

Time is divided into \( P \) steps of length \( \Delta t \) and indexed with an integer, \( p \), where an initial value problem for \( \hat{\chi}_n \) and \( \hat{\xi}_n \) is solved at each time step with initial conditions 

\[
\hat{\chi}_n(t) = \hat{a}_n(t) - \delta_{n,0}; \quad \hat{\xi}_n(t) = \hat{b}_n(t).
\]

Using the third-order Adams-Bashforth method, the time discrete form of Eqs. (10) takes the form, 

\[
\hat{\chi}_n^{p+1} = \hat{\chi}_n^p - \Delta t \left[ \frac{23}{12} (ab^2)_n^p - \frac{16}{12} (ab^2)_n^{p-1} e^{-\lambda_n \Delta t} \right] + \frac{5}{12} (ab^2)_n^{p-2} e^{-2\lambda_n \Delta t}.
\]

The values of the concentration at the new time step are found by inverting Eqs. (8), where \( t \) is replaced by the length of the time step, \( \Delta t \), and Fourier transforming to physical space. 

One advantageous aspect of this method is the use of the integration factors for the constant coefficient linear terms which correspond to the fastest times scales once the phenomena are sufficiently resolved in space. Once one submits to initial conditions which are spanned by the truncated Fourier space, the solution will be limited only by the number of bits used to represent the variables and the precision with which one can evaluate the exponential. The linear terms correspond to the fastest time scales due to the smoothing by viscosity, a rapid reaction of high wavenumber coefficients to the development of discontinuities in the variables caused by the nonlinear terms and nonequilibrium constraint. Thus when a problem is sufficiently resolved, discontinuities will form on a time scale longer than the e-folding time of the highest wavenumber spatial modes. Because the solution of these constant coefficient linear terms is manifestly implicit in the Fourier space this also leads to a scheme which is accurate when the numerical time step is long in comparison to an e-folding time of the highest-frequency spatial mode and stability is restricted by nonconstant coefficient linear or nonlinear terms.

IV. NUMERICAL STUDIES

In this section we present a comparison of a second-order finite difference method with the pseudo-spectral method in both one and two dimensions. In a spatial resolution study, we find that the speed of one-dimensional nonlinear traveling plane waves converges much more rapidly for the pseudo-spectral method than for the finite difference method. In two dimensions we find that the spectral method preserves the symmetry of the initial disturbance to a greater accuracy than the finite difference method.

A. One-dimensional wave speed and relaxation times

In one dimension in an infinite domain subject to a local arbitrary initial condition a number of countable disturbances will form on time scales of \( 1/(F + \kappa) \) if \( F \) and \( \kappa \) are chosen to be in a region of parameter space where plane nonlinear waves exist and the amplitude of the initial disturbance is large enough. Otherwise the state will relax to the trivial state on viscous time scales. In the former case these disturbances bounce off one another or begin propagating toward infinity. On time scales, \( T = c/L > 1/(F + \kappa) \), where \( c \) is the nonlinear wave speed and \( L \) is the width of the initial disturbance, the state will be composed of left and right traveling waves propagating away from each other toward infinity. In this section we measure these wave speeds by capturing and using a right traveling nonlinear wave as an initial condition.
For the purpose of discussion $F=0.02$, $\kappa=0.05$, $D_A=2.0 \times 10^{-5}$, and $D_B=10^{-5}$ are chosen. A typical right traveling wave is shown in Fig. 1. A richer set of behavior is found when different ratios of the diffusion coefficients and boundaries are considered as in Petrov, Scott and Showalter.\(^{21}\)

When a steady traveling wave solution to the equations of motion exists they take the form

$$a(x,t) = a(x-ct), \quad (13)$$

where $c$ is the wave speed. For this system of equations we hypothesize that the wave speed is unique—up to a change of sign—for a particular point in parameter, $F$-$\kappa$ space. When the solution takes this form the equations of motion reduce to

$$ca' + D_Aa'' - ab^2 + F(1-a) = 0,$$
$$cb' + D_bb'' + ab^2 - (F+\kappa)b = 0, \quad (14)$$

where $a$ and $b$ are now functions only of $\theta=x-ct$ and a prime, $'$, denotes differentiation with respect to $\theta$. In terms of a Fourier series of a right traveling wave, $c>0$,

$$a(x,t) = \sum \hat{a}_n(0) e^{i k_n (x-ct)} \quad \text{or} \quad \hat{a}_n(t) = \hat{a}_n(0) e^{-i k_n ct}, \quad \text{(15)}$$

and leads to

$$c = \hat{c}_n = \frac{1}{k_n} \tan^{-1} \left( \frac{\text{Im}[\hat{a}_n(t)\hat{a}_n^*(0)]}{\text{Re}[\hat{a}_n(t)\hat{a}_n^*(0)]} \right), \quad \text{(16)}$$

where Im and Re denote the real and imaginary parts respectively, and $*$ denotes the complex conjugate. The wave speed for Fourier coefficient, $\hat{c}_n$, is written to emphasize that a wave speed can be measured for each Fourier coefficient and that they all must be equal in order for Eq. (14) to be satisfied. To measure the wave speed we integrate the equations of motion, Eqs. (3), in time with an initial condition which is approximately the right traveling wave and thus an approximate solution to Eq. (14). The real and imaginary parts of the Fourier coefficients taken at two times, $T_c$, apart are used to measure the wave speed, $\hat{c}_n$, which depends on $n$ and on time until the solution relaxes to a steady state. Figure 2 shows a plot of $\hat{c}_n$ versus time for a typical run. Note that the individual instantaneous wave speeds relax as damped oscillators. Figure 3 shows the exponential decay of the magnitude of the error, $|\hat{\sigma}|=|\hat{c}_n(t_{\text{final}})-\hat{c}_n(t)|/\hat{c}_0(t_{\text{final}})$, in the wave speed versus time. Obviously the wave speed relaxes to a unique value for given resolution $\Delta t$, and initial condition $\Delta x$, sufficiently close to a solution of Eq. (14). The wave speed was measured the same way using the finite difference method, where it too relaxed to a unique wave speed. The relaxation time of the wave speed, $\tau_c$, is obviously a viscous relaxation time and might be estimated by

$$\tau_c = D_A L_R^2 (2\pi)^2 = O(50),$$

where $L_R$ is the full width at half maximum of the wave. For the chosen viscosities we estimate from the figures that the relaxation time is on the order of 200 dimensionless time units.

All runs that are used to estimate the wave speed were integrated for 5000 dimensionless time units to make sure that the error was less than the intrinsic errors in the wave.
speed due to spatio-temporal discretization errors. Visual inspection of the wave speeds measured for several Fourier coefficients and both the finite difference and pseudo-spectral methods revealed the wave speed is unique up to at least 10 digits of accuracy. Table I lists the measured wave speeds for different size time steps and a spatial resolution that will later be shown to have an error, \( \hat{\varepsilon}_1 \), less than \( 1.0 \times 10^{-11} \). The relative error in the wave speeds for Table I are estimated using the value obtained for \( \Delta t = 1.0 \times 10^{-3} \) as a reference and thus the errors, especially for the very small \( \Delta t \) runs, are underestimated. For the spatial resolution study we use a time step of \( \Delta t = 1.0 \times 10^{-2} \) except where stability limits of the finite difference method require an even smaller time step. This ensures that time stepping errors in the spatial resolution study are less than \( 1.0 \times 10^{-8} \).

Table II lists the measured wave speeds from the first Fourier coefficients for different resolutions. Table III lists the error, \( \hat{\sigma}_1 = \frac{\varepsilon - \hat{\varepsilon}_1}{\varepsilon} \), in the measured wave speeds using the highest resolution spectral calculation as a reference wave speed, where \( \varepsilon \) is the wave speed measured for the resolution \( N = 1204 \) of the pseudo-spectral code. Figure 4 is a graphical representation of the same table. We easily estimate the slope of the error in the finite difference method finding it to be \( 2.008 \) in good agreement with theory, \( \varepsilon \propto \Delta t^2 \). The error in the spectral method for the few points we have obtained indicates that the method does indeed fall off faster than any power of \( \Delta t \).

In the table we use the value of the wave speed calculated from the highest resolution run done with a Fourier method. This was done assuming that the Fourier method and the finite difference method will eventually converge to the correct value of the wave speed. This will overestimate the errors in the finite difference method if it is converging to a different wave speed. Thus we note that the error in the wave speed of the finite difference method calculated using the 4096 point run as a reference, changes the relative error in the 2096 resolution run from \( 1.631 \times 10^{-4} \) to \( 1.222 \times 10^{-4} \).
The latter estimate is obviously flawed in the same manner as the estimates in the errors in the temporal resolution study but we may still consider both estimates as good estimates and we have plotted the latter estimates in Fig. 4 and labeled them “Alternate Error of FD.”

From the error estimates we conclude that for a given error tolerance, a pseudo-spectral method with a lower resolution than the corresponding second-order finite difference method may be used. Once the phenomena are resolved the pseudo-spectral method is far cheaper than the second-order method may be used. Once the phenomena are resolved the pseudo-spectral method is far cheaper than the second-order finite difference method in terms of computational resources because a much lower resolution may be used.

B. Symmetry preservation in two dimensions

In previous studies of the Gray-Scott model with diffusion and a nonequilibrium constraint, one-dimensional plane waves and pattern formation from two-dimensional finite amplitude perturbations to the trivial state have been studied. A class of axisymmetric traveling waves also occurs and they are closely related to the plane wave solutions. However, these traveling waves sometimes occur in a region of the $F-k$ parameter space where plane waves do not occur and thus have a finite cutoff radius. Such waves reinforce the idea that spatially inhomogeneous waves are due to a balance of the algebraic nonlinear and nonequilibrium constraint terms with the diffusion terms which by their nature are related to the curvature of the variables representing the concentrations of the different chemical species.

These axisymmetric waves obey a one-dimensional form of Eqs. (3), where the Laplace operator is axisymmetric, $\nabla^2 \phi = \partial^2 \phi / \partial r^2 + 1 / r \partial \phi / \partial r$. Figures 5(a)-(d) show the evolution of an axisymmetric perturbation to the trivial state for both the spectral and finite difference schemes. The initial condition is

$$a = 1.0, \quad b = b_0 \exp\left(-\frac{(x-x_0)^2-(y-y_0)^2}{s^2}\right),$$

where $b_0 = 0.4, x_0 = y_0 = L/2, s = L/10$ and $L = 2.5$. The time step is chosen to be $\Delta t = 0.1$ although larger $\Delta t$ may be used and affects the magnitude of the phase speed but does not affect the axisymmetry of the wave. All runs were done using a grid that is $512 \times 512$ and spectral calculations were de-aliased to 255 coefficients. Figures 5(a),(b) show the results from the spectral scheme. Obviously, axisymmetry is well preserved until the disturbance reaches some critical radius where it oscillates and finally decays leaving the trivial steady state. Figure 5(c) shows the results from the finite difference method. This method has a tendency to form squares from axisymmetric initial conditions. For this run the behavior is manifested through the formation of patterns when the wave should decay to the trivial state. To emphasize the dependence of the formation of a square on the diffusion operator in the finite difference method, Fig. 5(d) shows the evolution of the same initial disturbance with a finite difference method where the diffusion operator has been rotated by 45°. We also note that because the diffusion operator is the only nonlocal operator, rotation of the diffusion operator by 45° decouples the problem into two problems because two uncoupled grids exist in this formulation. Some of the symmetry breaking properties of the finite difference method can be suppressed by going to high resolutions but as shown in the previous section the rate of convergence is slow relative to the pseudo-spectral method. Other finite difference operators, including higher order methods, might also be tried to suppress this behavior for particular situations. Circular truncation of the Fourier coefficients might be tried if one desires lower resolution pseudo-spectral calculations which preserve axisymmetry. At the same time care must be taken to de-alias the coefficients in the initial condition.

V. LINEAR INSTABILITIES

The nonlinear one-dimensional axisymmetric and plane waves of this system may be unstable to two-dimensional linear instabilities. Such instabilities are discussed by Hagedorn and Meron using a reaction diffusion model of the FitzHugh-Nagumo type. In addition, Pearson and Reynolds et al. discuss the instability and pattern formation of the system of reaction-diffusion equations we consider in this paper. Milton and Scott discuss the stability—planar, circular and spherical — of a related set of “cubic autocatalytic” reaction-diffusion equations including dependencies on the ratio of diffusion coefficient and the instantaneous curvature of the front. In this section, we present the equations for the linear eigenmodes of the nonlinear plane waves, some numerical evidence for their existence and a discussion of the linear instability and numerical evidence of the instability found in axisymmetric states. Studies of nonlinear plane wave instabilities were done on a domain, $1.5 \times 1.5$, with grid resolution of $256 \times 256$ while studies of axisymmetric phenomena were done on a domain, $2.5 \times 2.5$ with grid resolution $512 \times 512$. The time step was $\Delta t = 0.1$ in all cases presented.

For the linear instability theory of the nonlinear plane waves let $a$ and $b$ be composed of two parts,

$$a(x,y,t) = a_0(\theta) + a^1(\theta) e^{iy-\omega t}$$
$$b(x,y,t) = b_0(\theta) + b^1(\theta) e^{iy-\omega t},$$

where again, $\theta = x - ct$, the zeroth-order quantities, $a_0$ and $b_0$, are solutions to Eq. (14) and the first-order quantities, $a^1$ and $b^1$, are assumed small enough such that terms quadratic and cubic in these quantities may be neglected. The linear equations for the first-order quantities are,
FIG. 5. Color contour plots of the evolution of chemical concentrations of chemical A using the pseudo-spectral method unless otherwise noted for (a) an axisymmetric initial condition (b) same run as (a) but plotting chemical B, (c) an axisymmetric initial condition using a finite difference method, (d) same as (c) using a rotated Laplace operator, (e) a right traveling wave plus small two-dimensional perturbations, (f) an axisymmetric initial condition with a small perturbation consisting of 31 nonaxisymmetric modes, (g) same as (f) except that the small perturbation consists of one $m = 13$ nonaxisymmetric mode.
\[ \begin{align*}
\frac{\partial}{\partial \theta} + D_A \frac{\partial^2}{\partial \theta^2} - D_A a^2 - b_0^2 - F &= -2a_0b_0 \\
b_0^2 \frac{\partial}{\partial \theta} + D_B \frac{\partial^2}{\partial \theta^2} - D_B a^2 + 2a_0b_0 - F &= -\kappa
\end{align*} \]

(21)

with periodic or Dirichlet boundary conditions imposed in the \( \theta \) direction. Equation (21) describes a dispersion relation between \( \alpha \) and \( \omega \) with \( F \) and \( \kappa \) (and \( D_A \) and \( D_B \)) as parameters. If \( \alpha \) is a given real constant then these equations constitute an eigenvalue problem for \( \omega \), which is complex. When \( \alpha \) is nonzero and the imaginary part of \( \omega \) is positive definite the basic nonlinear wave is unstable to infinitesimal symmetry breaking linear perturbations. The eigenvalue, \( \omega_m \), which is the eigenvalue with the greatest value of the imaginary part for arbitrary wavenumber corresponds to the most unstable eigenmode and has a wavenumber, \( \alpha_m \). For a system, periodic in the tangential, \( y \), direction, there are one or possibly two most unstable eigenmodes with eigenvalue, \( \Im(\omega_m) \leq \Im(\omega_m) \), and they correspond to the most unstable wavenumbers, \( \alpha_{L_y} \), for a system with maximum periodicity length \( L_y \). For this system and the region of parameter space discussed the most unstable eigenmode has a finite wavelength. For the case, \( \alpha = 0 \), and parameters presented in the previous section, all eigenmodes are stable as apparent from the oscillation and decay of the error in the measured phase speed.

To measure the dynamics of the eigenmodes we define the modal mean squared concentration, \( \tilde{C}_{n,m} \), by the following relation

\[ \tilde{b}(x,y) = \sum_{n,m=0}^{\infty} \tilde{b}_{n,m} e^{i(kn+ikm)}, \]

(23)

where \( k_n = 2\pi n/L_x \) and \( k_m = 2\pi m/L_y \). In Fig. 6 we plot the square root of the tangential model mean squared concentration defined as \( \sqrt{\tilde{C}_{n,m}} = \Sigma_{n,m} \tilde{C}_{n,m} \). This plot shows the growth and decay of perturbations to the nonlinear wave with different tangential wavenumber, \( k_m \). The perturbation initially takes the form

\[ b_1(x,y) = \sum_{n=1}^{15} 10^{-8} \cos(k_n y)(1 + (x-x_0)) e^{-s(x-x_0)^2/8}. \]

(24)

where \( x_0 \) and \( s = L_x/10 \) are chosen such that the local disturbance decays to zero outside of the nonlinear wave. After a short adjustment period the coefficients, \( \tilde{C}_{n,m} \), grow or decay exponentially in time and are dominated by the dynamics of the most unstable/least damped eigenmode with wavenumber \( k_m \). The most unstable eigenmode for a system with

Periodicity length \( L = 1.25 \), parameters \( F = 0.02 \) and \( \kappa = 0.05 \) has a wavelength of \( \lambda = 0.25 \) and a growth rate of \( \Im(\omega) = 0.00379 \). Table IV gives the value of the growth rate of several eigenmodes measured from the data used to produce Fig. 6. These linear instabilities grow until their amplitudes are large enough for self-interaction and interaction with the basic nonlinear wave and other eigenmodes. A state characterized by irregular spatiotemporal patterns as discussed by Pearson13 usually results. Eigenmodes always excite tangential super- and sub-harmonic Fourier coefficients through nonlinear interaction but due to the small value of these harmonics they do not influence greatly the dynamics of the eigenmodes until they they are of sufficient amplitude. Figure 5 (e) shows this development from a time just before the perturbations begin to visibly interact with the one-dimensional axisymmetric nonlinear wave.

Axisymmetric nonlinear waves are also unstable to infinitesimal perturbations via linear symmetry breaking instabilities. We have not been able to derive an equation for a one-dimensional axisymmetric traveling wave of the form \( f(\theta) = f(r-ct) \). However, initially axisymmetric states propagate and remain axisymmetric and thus it is convenient to write the problem in polar coordinates and Fourier transform in the angular coordinate. In this context we consider the evolution of a state that consists of an axisymmetric state

**TABLE IV.** Table of grow rates of linear eigenmodes of the nonlinear plane wave with \( F = 0.02, \kappa = 0.055, D_A = 2.0 \times 10^{-3}, D_B = 1.0 \times 10^{-3}, L = 1.25, \Delta t = 1 \times 10^{-1} \).

<table>
<thead>
<tr>
<th>Mode number</th>
<th>Wave speed, ( \tilde{c}_n )</th>
<th>Growth rate</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
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<td></td>
</tr>
<tr>
<td>4</td>
<td>0.003323</td>
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<td>5</td>
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<td>6</td>
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<td></td>
</tr>
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<td>7</td>
<td>0.003266</td>
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</tr>
<tr>
<td>8</td>
<td>0.002193</td>
<td></td>
</tr>
</tbody>
</table>
plus a small nonaxisymmetric perturbation characterized by a mode number, \( m \), and a time and radially dependent amplitude,
\[
\begin{align*}
    a(r, \phi, t) &= a_0(r, t) + a_1(r, t) e^{i m \phi}, \\
    b(r, \phi, t) &= b_0(r, t) + b_1(r, t) e^{i m \phi}.
\end{align*}
\]

Neglecting terms second order in the perturbation quantities, the axisymmetric component obeys the following equation of motion,
\[
\begin{align*}
    \frac{\partial a_0}{\partial t} &= D_A \nabla^2 a_0 - a_0 b_0^2 + F(1 - a_0), \\
    \frac{\partial b_0}{\partial t} &= D_A \nabla^2 b_0 + a_0 b_0^2 + (F + \kappa) b_0, \\
\end{align*}
\]
where \( \nabla^2 = \partial^2/\partial r^2 + 1/r \partial/\partial r \). The first-order equations for the perturbation are,
\[
\begin{align*}
    \frac{\partial a_1}{\partial t} &= D_A (\nabla^2 - m^2) a_1 - (2a_0 b_0 b_1 + b_0^2 a_1) - F a_1, \\
    \frac{\partial b_1}{\partial t} &= D_A (\nabla^2 - m^2) b_1 + (2a_0 b_0 b_1 + b_0^2 a_1) - (F + \kappa) b_1.
\end{align*}
\]

So Eq. (27) is a linear equation for the nonaxisymmetric component of the initial state driven by the nonlinear equation for the axisymmetric component. For the parameters we have explored the nonaxisymmetric component grows for some mode numbers and thus the axisymmetric part is linearly unstable to symmetry breaking instabilities.

This instability is important, as it allows infinitesimal perturbations to grow in regions of \( F - \kappa \) parameter space where plane nonlinear waves do not exist. It is also easy to introduce finite amplitude axisymmetric perturbations plus a small amount of random noise to a system. The axisymmetric perturbation forms and propagates while at the same time amplifying nonaxisymmetric noise. Figure 5(f) shows the development of a linear instability on an axisymmetric nonlinear wave. To the axisymmetric initial condition, Eq. (18), a small perturbation is added of the form:
\[
b_1(x + x_0, y + y_0) = \sum_{n=1}^{31} 10^{-5} \cos(m \theta + \phi) r e^{-r^2/s^2},
\]
where \( x_0 \) and \( y_0 \) are the point of symmetry, and \( s = L/10 \) are chosen so that the perturbation is located within the initial axisymmetric disturbance. These linear perturbations grow and eventually lead once again to a state characterized by the formation of irregular patterns. Again there will be a most unstable mode, \( m \), but in this case \( m \) will depend on the radius of the wave and thus may not be a unique function of \( F \) and \( \kappa \). To emphasize that the method resolves these modes, Fig. 5(g) shows the development of the same axisymmetric initial state but the small perturbation consists of a single, \( m = 13 \), mode with amplitude equal to \( 10^{-4} \). These numerical experiments clearly show that certain fundamental quasi-steady nonlinear one-dimensional plane and axisymmetric waves are linearly unstable to two-dimensional perturbations.

VI. DISCUSSION

We have implemented a semi-implicit pseudo-spectral Fourier method to solve the equations for the reaction-diffusion form of the Gray-Scott model with a nonequilibrium constraint finding that such a method is quite powerful for the exploration of fundamental phenomena which occur in this system. This method is based on expanding the concentrations of the chemicals in a Fourier series and solving the equations for the Fourier coefficients using an integrating factor where the nonlinear terms are fully de-aliased and act as time-dependent forcing functions.

This method, in comparison to a standard second-order finite difference method, has advantageous numerical stability properties, a high rate of convergence and excellent symmetry preserving properties. Because the pseudo-spectral method uses an integration factor it is manifestly implicit in the diffusion and other constant coefficient linear terms. The convergence rate of the finite difference method is, as expected, proportional to \( \Delta x^2 \) while the pseudo-spectral method converges faster than any power of \( \Delta x \). For the pseudo-spectral method resolved axisymmetric initial conditions remain axisymmetric until interactions due to periodic boundary conditions break the symmetry, while the same initial conditions tend to form square structures and patterns for the simple finite difference method.

These numerical properties allow the accurate calculation of one-dimensional wave speeds and the exploration of several phenomena that are sensitive to noise from initial conditions and numerical error. Annular structures are found but form only in very low noise environments. These axisymmetric traveling waves may exist in regions of \( F - \kappa - D_A - D_B \) parameter space where plane waves are not possible and thus have a finite radius. Both the plane and axisymmetric waves are linearly unstable to two-dimensional disturbances with finite most-unstable wavenumbers. These linear instabilities provide a route to a pattern forming state from infinitesimal perturbations.

VII. NOTE ON PARALLELIZATION

We would like to note that the pseudo-spectral method is an intrinsically parallelizable method except for the fast Fourier transform which is usually one of the top priorities for optimization on any computer system, including parallel computers, due to its diverse and important range of applications. We would also like to note that any finite difference method will need an implicit solver for the viscous term if it is to compete with the Fourier method at high resolution and thus will also require some global communication patterns.

ACKNOWLEDGMENTS

The base support for COAPS is from the Office of Naval Research, Physical Oceanography Section. Wesley B. Jones

W. B. Jones and J. J. O'Brien: Methods and instabilities in fronts
APPENDIX: DE-ALIASING CUBIC NONLINEARITIES

To integrate the equations of motion in Fourier space, we must evaluate the Fourier coefficients of the nonlinear term. We use an inverse discrete Fourier transform (DFT) to transform \( \hat{a}_n \) and \( \hat{b}_n \) to physical space where the multiplication is performed, and then again use the DFT to determine \( \hat{c}_m \). If this procedure is not done carefully, the result, \( \hat{c}_m \), will be aliased. We remove this aliasing error by padding the DFT. The argument presented closely follows that given in Canuto et al.\textsuperscript{20} for de-aliasing a second-order polynomial.

Consider that we represent \( a(x) \) and \( b(x) \) at \( M \times N \) discrete points as a Fourier series,

\[
\begin{align*}
\hat{a}_m & = \sum_{j=-M/2}^{M/2-1} \hat{a}_m e^{i(2\pi jm/M)}, \\
\hat{b}_m & = \sum_{m=-M/2+1}^{M/2-1} \hat{b}_m e^{i(2\pi jm/M)},
\end{align*}
\]  

(A1)

where the coefficients \( \hat{a}_m \) and \( \hat{b}_m \) have been padded with zeros:

\[
\hat{a}_m = \begin{cases} 
\hat{a}_m, & |m| < N/2, \\
0, & \text{otherwise}.
\end{cases}
\]  

(A2)

The nonlinear term is found on the same set of points and the coefficients of the nonlinear terms are found from the DFT,

\[
c_m = a_j b_j^2, \quad \hat{c}_m = \frac{1}{M} \sum_{j=0}^{M-1} c_j e^{-i(2\pi jm/M)}.
\]  

(A3)

The orthogonality relation of the DFT is

\[
\frac{1}{M} \sum_{j=0}^{M-1} e^{i(2\pi jm/M)} = \begin{cases} 
1, & m = Mn, \ n = 0,1,2,\ldots, \\
0, & \text{otherwise},
\end{cases}
\]

and results in the following relation,

\[
\tilde{c}_m = \frac{1}{n+p+q-M} \sum_{n+p+q-M=r} \hat{a}_n \hat{b}_p \hat{b}_q, \quad r=0,\pm 1,\pm 2,\ldots.
\]  

(A4)

All terms that include \( r > 0 \) are aliased terms. The trick is to choose \( M \) greater than \( N \) such that all terms with \( r > 0 \) are identically zero. The minimum condition on \( M \) is

\[
4(\frac{N}{2} - 1) < M \quad \text{or} \quad M \geq \frac{N}{2} - 1.
\]  

(A5)

Thus to determine the Fourier coefficients of a cubic nonlinearity from the Fourier coefficients of the individual variables, one applies the Fourier transform from a Fourier space with \( N \) coefficients to physical space using a DFT with \( M > N/2 - 1 \) coefficients, performs the multiplication there, and transforms back to Fourier space.

\[\text{References}\]


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