Complex Patterns in Reaction-Diffusion-Systems

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Summary:

Complex patterns in reaction-diffusion systems resist a complete characterization by simple geometric parameters, while simple patterns are well described by a few quantities like speed, wavelength, frequency and symmetry. Among complex patterns we can distinguish between I) patterns, wherein many simple structures compete and interact, II) irregular, disordered and chaotic patterns and III) complicated patterns created by external perturbations. These three cases give the outline of this thesis. We study nonlinear partial differential equations by means of numerical integration, perturbation theory, continuation methods and numerical stability analysis.

We obtain the following main results:

- **Pattern competition:**
  Interfaces between coexisting patterns show phenomena like wavenumber selection and interface locking. Stable drifting pattern domains are found far from the onset of pattern formation.

- **Interaction of pulses and spirals:**
  Interaction between coherent structures can be studied in an exact manner by numerical stability theory. Results from analytic kinematical theories are verified in some situations. In one dimension, bound states of pulses are found for anomalous dispersion. In two dimension, spirals near a mirror wall display boundary induced drift. Experiments in several chemical reactions are modelled and explained in view of these results.

- **Transitions to spatiotemporal chaos:**
  Spatiotemporal chaos emerges if pulses and spirals become linearly unstable or undergo a bifurcation. Three scenarios for pulse bifurcations and four scenarios for spiral instabilities have been identified. The transition from phase to defect chaos in one-dimensional oscillatory media is explained and the nucleus for defect creation is identified.

- **Characterization of spatiotemporal chaos:**
  Spiral-defect chaos in a two-dimensional reaction-diffusion systems is characterized by a statistics of topological defects. Correlations between defects strongly influence the fluctuations of the defect number around its mean value. A gradual transition from a defect liquid to a defect gas is described.
- **Anisotropic patterns:** Theories for the formation of square shaped waves and front propagation in anisotropic media are presented. A new pattern, stratified spatiotemporal chaos, is discovered.

- **Heterogeneous media:** Various patterns in heterogeneous media are reproduced. Homogenization is applied to reaction-diffusion media.

- **Noisy pattern formation:** Stochastic models explain phenomena in biological systems like intracellular calcium waves and myxobacterial aggregation.
1 Introduction: Reaction-Diffusion-Systems in Physics, Chemistry and Biology

1.1 What are Complex Patterns that We should be Mindful of Them?

Spontaneous pattern formation under natural and laboratory conditions is a trademark of systems far from thermodynamic equilibrium. These systems are typically subject to a constant throughput of matter and energy and can be classified as open and dissipative systems. In closed systems patterns that may emerge initially typically decay in the long run and the systems approach a featureless, spatially homogeneous state - thermodynamic equilibrium. This is in line with the demands of the second law of thermodynamics: the entropy in a closed system increases until equilibrium is reached. Open systems, however, are not subject to the constraint of the second law - they can export entropy to their surroundings by transforming energy of high quality into energy of low quality, typically heat. The latter process is known as dissipation.

Theoretical research has initially focussed on the question under which nonequilibrium conditions a system switches from a homogeneous state to a pattern. These transitions are known as instabilities or bifurcations and can be classified in a spirit similar to equilibrium phase transitions [1]. For example, one can distinguish between continuous (supercritical) and discontinuous (subcritical) instabilities. The emerging pattern may be stationary (Turing instability) or dynamic (Hopf instability) as well as of periodic or localized nature. In one spatial dimension, periodic stationary pat-
terns are typically stripes, while the range of dynamic patterns comprises traveling and standing periodic waves. In higher dimensions, periodic patterns display various symmetries, e.g. in two dimensions parallel stripes, hexagons and square patterns are known. Localized patterns include traveling and stationary fronts and pulses in one dimension and spots and spheres in two resp. three dimensions. An interesting combination between a localized and a periodic pattern is given by a rotating spiral, that has a well defined pointlike center of rotation (core), from which periodic waves are emitted in all radial directions. So far we have mentioned only simple patterns. These patterns are well described by a few quantities like speed, wavelength, frequency and symmetry. A gallery of simple patterns is shown in Fig. 1.1. In three dimensions, spiral cores in the plane are transformed into lines that are called filaments. If the filament forms a straight line, one speaks of a scroll wave. A scroll ring, where the filament forms a closed loop, is shown in the lower panel of Fig. 1.1. Experimental studies of three dimensional structures are much more difficult due to imaging problems. In particular no controlled experiments about complex patterns have been performed so far. This is why in the remainder of this thesis, we confine ourselves to one- and two-dimensional pattern formation.

While many experimental and theoretical studies have been performed on the properties of these simple structures, considerable less is known about complex patterns that resist a complete characterization by simple geometric parameters. Such patterns are, however, typically observed in nature. Observation of isolated simple patterns usually involves intriguing experimental techniques, while spontaneous processes yield either typically at least a superposition of simple patterns or a perturbed by external and internal noise sources. Among complex patterns we can distinguish between I) patterns, wherein many simple structures compete and interact, II) irregular, disordered and chaotic patterns and III) complicated patterns created by external perturbations. These three cases form roughly the structure of this thesis.

Before we go any further, let us consider some illustrative examples of complex patterns that emerge in nature as well as in laboratory experiments. One of the most frequent patterns in reaction-diffusion media are rotating spiral waves. Typically more than one spiral wave form spontaneously. An example of three interacting spiral waves observed experimentally [2] in the catalytic reduction of NO with CO on a platinum surface is shown in the left panel of Fig. 1.2. In the long run, the three spiral repel each other and increase their mutual distance. While the dynamics of the individual spiral is well described by the (fast) rotation frequency and the spiral geometry, the
d=1

spatial profile $u(x)$

(v=0 also possible)

---

d=2

isolines $u(x,y) = u_0$

hexagons

target

spiral

stripes

---

d=3

front position

scroll-ring

---

Fig. 1.1: Examples for simple patterns in one, two and three dimensions
interaction of spirals happens on a slow time scale and requires long observation times. The interaction of spirals in most media is an unsolved theoretical problem. More details on spiral interaction can be found in Chapter 2.

An even more complicated situation arises in the aggregation of slime molds, see right panel of Fig. 1.2. There, the concentration of the chemoattractant cAMP forms rotating spiral waves, that divide the plane into subdomains. From each of these subdomains, all cells tend towards the respective spiral center due to chemotaxis and gather there before the next stages of aggregation happen. Successful aggregation requires a certain critical amount of cells. Therefore, it is crucial that not too many spirals survive the initial stage where cells are still distributed almost homogeneously in the plane. Indeed, experiments show a drastic reduction of the initial number of spirals by mutual annihilation of spirals with opposite sense of rotations [4]. This „coarsening” process of spiral domains has been reproduced in caricature models [5] and realistic equations [3] and provides an example for the need of pattern competition in a biological system.

Spatiotemporally chaotic patterns provide a second type of widely encountered
Fig. 1.3: Examples for spatiotemporal chaos in one-dimension (space-time plot, left) and two dimensions (snapshot, right).
complex structures. The corresponding phenomena have been studied in many model systems and were verified in laboratory experiments. Two „experimental” examples in excitable media in one resp. two dimensions are shown in Fig. 1.3. The left panel shows pigment pattern on a sea shell and a corresponding simulation with a qualitative model by Meinhardt [6]. Since shells are growing layer by layer, the pigment pattern is often interpreted as a space-time diagram of an effectively one-dimensional system. In this example the pattern obviously contains many different lengthscales and cannot be characterized by a typical wavelength anymore. A quantitative analysis of the onset of spatiotemporally chaotic pattern formation has first been achieved in laboratory experiments in a quasi-two-dimensional chemical reaction, namely the Belousov-Zhabotinsky reaction [7]. A snapshot of a chaotic patterns from these experiments is shown in the right panel of Fig. 1.3. The image is recorded just above the transition from regular spirals to chaos. In the upper right quarter of the frame one still sees some remainder of regular spiral structures. In this thesis, we study transitions from regular to chaotic dynamics in one- and two-dimensional reaction-diffusion media, see Chapter 3. Finally, complexity may be forced by intrinsic or external perturbations to the homogeneity of the medium. A suitable experimental system is the light-sensitive Belousov-Zhabotinsky reaction [8]; an illustration of experiments with periodic illumination of the right half of the medium is shown in Fig. 1.4. The time-periodic forcing with light changes the pattern from perfectly periodic waves to a disordered labyrinthine pattern. A large variety of other patterns have been obtained upon changes of the frequency and amplitude of the illumination. The example shows that pattern formation can be influenced and to a certain extend „controlled” by external manipulation of the self-organization processes. Many other strategies lead to similar results. Various examples are reported in the Chapters 4 and 5 of this work.

To summarize, in Chapter 2 we study complexity arising from competition or interaction of simple patterns. Intrinsic instabilities and bifurcations may prevent the formation of any simple pattern. Complex patterns that arise in such a situation are often irregular in space and time and fulfill the criteria for spatiotemporal chaos or weak turbulence. Several case studies of these phenomena are presented in Chapter 3. Moreover, the breaking of the translational and rotational symmetries by anisotropy and heterogeneities described in Chapter 4 is a potential source of complex pattern formation. Most pattern forming media are well approximated by deterministic models; however patterns may also appear in fluctuating, stochastic media as is reported in Chapter 5.
Fig. 1.4: Pattern formation in the Ruthenium catalyzed Belousov-Zhabotinsky reaction under homogeneous conditions (left) and under periodic illumination (right), after [8].
Experiments on pattern formation have been conducted in thermal and electrohydrodynamical convection as well as in a large variety of reaction-diffusion systems in physics, chemistry and biology. The latter type is typically modelled by nonlinear coupled partial differential equations that describe the coupling between local changes in concentrations by „reactive” processes and simple diffusive transport of the „reactands” involved. Examples for reaction-diffusion systems in physics are gas-discharge systems [9] and semiconductors [10, 11]. Many chemical reactions show reaction-diffusion patterns [12]. In biology, reaction-diffusion behavior is encountered on the cellular level in action potential propagation [13] and intracellular calcium waves [14] as well as in colonies of microorganism like the slime mold Dictyostelium Discoideum [15]. While the physical and chemical processes underlying these phenomena are quite diverse, the basic principles and the form of the arising models are often surprisingly similar. In particular, it makes sense to study basic phenomena first in simple model equations like the FitzHugh-Nagumo equations [16, 17]. Such results supplement the study of realistic models based on the knowledge of elementary processes and help to identify common properties and potentially universal behavior in pattern forming systems. In the next sections, we will give an outline and a short history of reaction-diffusion systems, introduce important experimental applications and discuss model equations that are relevant to the remainder of this thesis. Simplified models allowing for analytical treatment of complex pattern formation phenomena are introduced. Finally, we will give a brief overview of the mathematical and numerical methods used in this work.

1.2 A History of Reaction-Diffusion Systems

The most remarkable property of reaction-diffusion systems is spontaneous formation of a great variety of patterns. These structures do not offer only aesthetical pleasure, but sometimes also provide efficient means of communication and signal transmission. In order to so, a reaction-diffusion medium has to be sensitive to small stimuli from the enviroment and must be able to propagate them in a reliable and fast fashion. Excitable media are particular well suited for that purpose. They have a stable rest state and a threshold. Perturbations larger than the threshold may cause a large response, while small perturbations and noise decay immediately. Superthreshold perturbations lead via diffusion to propagation of fast reaction-diffusion waves that transmit information in a reliable fashion. A second more controversial application of reaction-diffusion systems is their ability to form stationary periodic patterns (Turing structures). Such structures
may play an important role in morphogenesis and the development of structures in living organism.

Historically, the earliest model for a two-dimensional excitable medium is a cellular automaton model for the propagation of electrical activity in heart muscle [19] proposed by Wiener and Rosenblueth already in 1946. The field of reaction-diffusion systems as we know them can to a large extent traced back to two quite different landmark papers published in 1952. British mathematician Alan Mathison Turing considered the „Chemical Basis of Morphogenesis” [18] and showed that the interplay of nonlinear reaction and diffusion transport may lead to sustained stationary concentration patterns, henceforth often called „Turing structures”.

The first example of an excitable medium derived from underlying physico-chemical processes has been provided in 1952 by british physiologists A. L. Hodgkin and A. F. Huxley. They derived a set of ordinary differential equations neglecting spatial variations from measurements of ionic currents at the membrane of the squid giant axon [13]. Their nobelprizewinning effort is still considered the „most successful model in physiology” [20]. The resulting Hodkin-Huxley model accounts for the dynamics of action potentials in neurons. The equations describe the dynamics of the membrane voltage $V$ as a function of the ionic currents of sodium and potassium and read

$$
egin{align*}
\frac{c_m}{dt} V &= -g_K n^4 (V - V_K) - g_{Na} m^3 h (V - V_{Na}) - g_L (V - V_L) + I_{app} \\
\frac{dm}{dt} &= \alpha_m (1 - m) - \beta_m m \\
\frac{dn}{dt} &= \alpha_n (1 - n) - \beta_n n \\
\frac{dh}{dt} &= \alpha_h (1 - h) - \beta_h h,
\end{align*}
$$

where $m$, $h$ and $n$ denote the gating variables for sodium channel activation and deactivation and for potassium channel activation, respectively. The membran voltage $V$ feeds into the time constants $\alpha_i, \beta_i$ for channel activation and deactivation.

The concept of an excitable medium described by continuous variables has found many applications in pattern forming chemical and biological systems [21]. Since the Hodgkin-Huxley equations and many of the subsequently developed models for membrane potentials [20] are coupled nonlinear ordinary differential equations, they have largely resisted analytical treatment and have been mostly studied numerically. A largely simplified version has been derived by R. FitzHugh and J. Nagumo in the early
It is known as the FitzHugh-Nagumo model [16] and reads
\[
\begin{align*}
\frac{du}{dt} &= -\frac{u^3}{3} + u - v = f(u, v) \\
\frac{dv}{dt} &= \epsilon(u - \gamma v + \beta) = \epsilon g(u, v).
\end{align*}
\] (1.2)

Originally, the activator \(u\) is derived from the voltage \(V\) of the Hodgkin-Huxley equations, while the inhibitor \(v\) represents a caricature of the slow gating variable \(n\) [20]. It will be the basis of generic model approaches presented in the later parts of this thesis.

If one allows for spatial variation of the defining variables one can simply add transport by diffusion and obtains a coupled set of *nonlinear partial differential equations*
\[
\begin{align*}
\partial_t u &= f(u, v) + D_u \Delta u \\
\partial_t v &= \epsilon g(u, v) + D_v \Delta v.
\end{align*}
\] (1.3)

If one intends to use the FitzHugh-Nagumo model as a description for propagating action potentials, inhibitor diffusion has to be neglected, i.e. \(D_v = 0\). The spatiotemporal dynamics of these equations is governed by the control parameters \(\beta, \gamma\) and \(\epsilon << 1\) and the diffusion constants \(D_u\) and \(D_v\). For convenience, we use the notation \(\partial_t u\) for the more common \(\partial u / \partial t\) in all reaction-diffusion equations in this and subsequent chapters. The variables are often specified as (fast) activator \((u)\) and (slow) inhibitor \((v)\). This terminology goes back to the work of Gierer and Meinhardt [22], which in turn has been developed on the basis of Turing’s ideas. A nice feature of the FitzHugh-Nagumo model is that it contains both the Turing patterns and the excitable medium as special cases depending on the tuning of the parameters. Requirements for excitability are fast activator dynamics \((\epsilon << 1)\) and diffusion \((D_u / D_v > 1)\) as well as a small, positive excitation threshold \((0 < |\beta - 2\gamma/3 + 1| << 1)\). Turing structures on the other hand require at least slow inhibitor diffusion \((D_u / D_v < 1)\). The FitzHugh-Nagumo equation will appear in various modifications in this thesis. Many real systems like action potential propagation [23], calcium waves [20] or catalytic CO oxidation [24] do not follow the simple linear relation for the inhibitor production in the FHN equations, but rather more complicated nonlinear dependencies. A computationally more efficient version of the FitzHugh-Nagumo model has been provided by Barkley [25] and modified by Bär and Eiswirth [26] to study spatiotemporal chaos. Its general form reads:
\[
\begin{align*}
\partial_t u &= \frac{1}{\epsilon} u(1 - u)(u - \frac{b + v}{a}) + \partial_x^2 u, \\
\partial_t v &= h(u) - v.
\end{align*}
\] (1.4)
Excitable conditions are realized for \( a < 1 \), \( b \) positive and small and \( 0 < \epsilon << 1 \). Barkley’s original version uses a linear inhibitor production \( h(u) = u \). For excitable conditions, the medium then has a single homogeneous fixed point \((u, v) = (0, 0)\) like the original FHN model. We shall refer to this situations as excitable medium of type I. The modification of Bär and Eiswirth introduces a nonlinear, cubic function \( h(u) \) for the inhibitor production, that leads to additional unstable homogeneous fixed points. We will refer to this situation as excitable medium of type II. The simple change in the inhibitor production leads to interesting new physics including anomalous dispersion for pulse trains resp. attractive pulse interaction (see Chapter 2) and appearance of spatiotemporal chaos via pulse backfiring in one and spiral breakup in two dimension (see Chapter 3). A second thread are extensions of the standard FitzHugh-Nagumo model that cover topics like the impact of a third variable or nonlocal coupling and geometrical constraints (see Chapter 1) as well as diffusional anisotropy and the influence of heterogeneities (see Chapter 4).

In the course of the 1960s, the interest in „dissipative structures” in chemical systems started to grow. As a simplification of Turing’s model, Lefever, Nicolis and Prigogine suggested the following reaction scheme

\[
\begin{align*}
A & \rightarrow X; B + X \rightarrow Y + D \\
2X + Y & \rightarrow 3Y; X \rightarrow E
\end{align*}
\]

where the concentrations of \( A \) and \( B \) are used as control parameters and fixed at constant values \( a \) and \( b \), respectively.

The corresponding reaction-diffusion model is widely known as the „Brusselator” and reads

\[
\begin{align*}
\partial_t u & = a - (b + 1)u + u^2v + D_u \nabla^2 u \\
\partial_t v & = bu - u^2v + D_v \nabla^2 v,
\end{align*}
\]

(1.5)

where \( u(\bar{x}, t) \) and \( v(\bar{x}, t) \) denote the concentrations of \( X \) and \( Y \). All rate constants have been set to unity. The Brusselator allows for oscillations, if \( a > a_c = b^2 + 1 \) in the case \( D_u/D_v > 1 \), and for a Turing instability, if \( D_u/D_v < 1 \). It has been used as a prototype model for the emergence of „dissipative structures” by Prigogine,
Nicolis and colleagues [27, 28], by Kuramoto [29] and many others. It may serve here also as a simple example for a strategy widely used in the modeling of chemical and biological reaction-diffusion systems. First, identify the kinetic scheme for a particular system, second write down the corresponding set of differential equations, third add the relevant transport processes (diffusion, convection etc.) and last but not least look out for multistability as well as dynamic, oscillatory resp. pattern forming instabilities. Excitable media usually appear near oscillatory regimes. Multistable systems should exhibit fronts between stable states that typically travel with constant speed and shape. Since the arrival of the Brusselator, this strategy has been applied to many systems in homogeneously and heterogeneously catalysed chemical reaction [12] as well as in biochemical and biological systems [30].

For two variable activator-inhibitor models, this analysis can often be done by simply considering the nullclines. These curves are the zeros of the reaction part in the twodimensional phase space. Their intersections immediately give the homogeneous steady states of the model. For excitable media, typically a Z-shaped activator nullcline intersects the monotonous inhibitor nullcline. Stable states are obtained for intersection on the outer branches of the Z-nullcline, intersection on the inner branch of the Z correspond to unstable fixed points as long as the activator dynamics is muched faster than the inhibitor dynamics. Examples for nullclines in excitable, oscillatory and bistable media are shown in the left column of Fig. 1.5 including typical trajectories in the phase plane and the corresponding wave profiles (right column). All curves have been obtained with the modified version of the Barkley model (1.4), the parameters in the top row correspond to a type II excitable medium.

In parallel to the first studies of the Brusselator model, experimental observations of dissipative structures in the form of target patterns [31] and spiral waves [32] have been reported with the Belousov-Zhabotinsky (BZ) reaction. The BZ reaction is the oxidation of malonic acid and involves more than 100 chemical species. Nevertheless, Field, Körös and Noyes extracted a core mechanism of the reaction that has become known as the Oregonator model [33]; it explicitly includes only three species. Until the early 1990s, more and more details of spiral dynamics in the BZ reaction have been investigated [34]. However, many results until then have been limited due to the use of ,,closed” reaction vessels. A major breakthrough has been the design and use of open reactors both in homogeneously and heterogeneously catalyzed reactions (see Fig. 1.6). They allow for steady supply of educts and removal of products, thus maintaining constant concentrations of key species and keeping the system far away from chemical
1.2 A History of Reaction-Diffusion Systems

Fig. 1.5: Nullclines of the reaction ODEs (thick lines) of Eqs. (1.4) and phase plot of wave solutions (thin lines) are shown on the left. Spatial profiles of wave solutions appear on the right. (a) Excitable medium $\epsilon = 0.025$, $a = 0.75$, $b = 0.07$, is characterized by a single stable fixed point and solitary pulse solutions. (b) Bistable medium $\epsilon = 0.025$, $a = 1.2$, $b = 0.1$, is characterized by two stable fixed points and front solutions. (c) Oscillatory medium $\epsilon = 0.025$, $a = 0.5$, $b = -0.05$, is characterized by no stable fixed points and travelling waves as well as homogeneous oscillations.
equilibrium.

The original Turing structures have been discovered in the Chlorid-Iodid-Malonic-Acid (CIMA) reaction [35]. Later on, the CIMA reaction was investigated in extended two-dimensional domains [36]. The second half of the 1990s has then seen the discovery of further structures, labyrinthine patterns [37] and replicating spots [38]. A second exciting line of research in pattern-forming chemical systems originated from the study of reaction on catalytic surfaces [39, 40]. In particular, the CO oxidation on Pt(110) revealed a huge variety of qualitatively different patterns. Catalytic reaction offer various advantages: they can be operated under a wide range of external conditions regarding pressure and temperature, they are truly two-dimensional and last but not least, they introduce new aspects into pattern formation including global coupling and anisotropy. For reviews on patterns in catalysis, see [42, 41].

Another important field where reaction-diffusion processes play a prominent role are nonlinear waves and pattern formation in biological systems. Pioneering experiments in aggregating slime mold colonies revealed spiral waves of chemoattractant in the early aggregation stage [43]. By now the whole cycle of aggregation and the spatial patterns associated with its stages have been thoroughly studied, for reviews see [44, 15]. Another more recent example of reaction-diffusion behavior in biology are intracellular calcium waves [14]. For all these systems, reaction-diffusion models have been derived. Surprisingly, the simple activator-inhibitor picture as well as the concept of an excitable medium could be applied to most of these examples.

1.3 Mathematical Concepts and Numerical Methods

Consider a general $N$-component reaction diffusion problem characterized by the vector $\vec{U}$ of time and space-dependent concentrations $U_i(x,t)$:

$$
\partial_t \vec{U}(x,t) = \vec{F}(\vec{U}(x,t), \vec{\mu}) + \vec{D} \nabla \vec{U}(x,t),
$$

(1.6)

where $\vec{\mu}$ contains the control parameter(s) $\mu_i$ and $\vec{D}$ is the diffusivity tensor which is assumed to be diagonal (no cross-diffusion) and independent of space and concentrations (linear diffusion). These equations have to be supplemented by boundary conditions.
Fig. 1.6: Sketch of open reactor types used in (a) homogeneous catalysis - continuously fed unstirred reactor (CFUR) - and (b) in heterogeneous catalysis
1.3.1 Onset of Pattern Formation and Weakly Nonlinear Theory

The onset of pattern formation is usually associated with the linear instability of homogeneous stationary solutions in Eqs. (1.6). These solutions are defined by \( F(U_S) = 0 \) and can be found with Newton’s method. Next, one has to consider small perturbations \( u \) around this solution: \( u(x,t) = U(x,t) - U_S \). Plugging this expression into the original equations and expanding to first order in \( u \), one obtains

\[
\frac{\partial_t u}{\partial t} = \mathcal{L}(U_S, \mu)u + \mathcal{O}(u^2) \tag{1.7}
\]

where \( \mathcal{L} \) is a \( N \times N \) matrix, the elements of which are given by operators \( \mathcal{L}_{ij} = \frac{\partial^2 F_i}{\partial U_j(U_S)} + D \delta_{ij} \). In one dimension and with the ansatz \( u(x,t) \propto \exp(\omega_n t - kx) \), one obtains an eigenvalue problem

\[
\mathcal{L}(U_S, \mu) \Phi_n = \omega_n \Phi_n \tag{1.8}
\]

where \( \omega_n \) are the growth rates of the corresponding modes \( \Phi_n \). The steady state \( U_S \) is called linearly stable, if all \( \omega_n \) fulfill \( \text{Re} \omega_n \leq 0 \). An instability or bifurcation is found at \( \mu = \mu_C \) from the condition \( \text{Re} \omega_n(\mu_C) = 0 \). Note, that \( \omega_n(\mu_C) \) depends on the wavenumber of the perturbation \( k \) and may also have an imaginary part. The eigenmodes have to be consistent with the boundary conditions of the original problem not explicitly stated here. The stability properties often depend strongly on the boundary conditions used. Thus the instability can be associated with a critical wavenumber \( k_C \) and may also have a nonzero frequency \( \Omega_C = \text{Im} \omega_n(\mu_C) \). The simplest case of a pattern forming instability is the Turing bifurcation with \( k_C \neq 0 \) and \( \Omega_C = 0 \). With the ansatz \( U(x,t) = U_S + \Phi_n^C \mathcal{R}(x,t) e^{ik_C x} + c.c. \) and performing a weakly nonlinear analysis we obtain the real Ginzburg-Landau equation (RGLE) after proper rescaling of space and time

\[
\frac{\partial_t \mathcal{R}}{\partial t} = \eta \mathcal{R} + \frac{\partial^2 \mathcal{R}}{\partial_x^2} - |\mathcal{R}|^2 \mathcal{R} \tag{1.9}
\]

The amplitude \( \mathcal{R} \) of the pattern is complex, while the coefficients of the terms in Eq. (1.9) are real. For an oscillatory instability with \( k_C = 0 \) (Hopf bifurcation), the ansatz \( U(x,t) = U_S + \Phi_n^C A(x,t) e^{ic_3 t} + c.c. \) yields the famous complex Ginzburg-Landau equation

\[
\frac{\partial_t A}{\partial t} = \rho A + (1 + ic_1)\frac{\partial^2 A}{\partial_x^2} - (1 + ic_3)|A|^2 A \tag{1.10}
\]

Additional rescaling of the complex amplitudes \( A, \mathcal{R} \) allows to set the linear coefficients \( \eta, \rho \) to unity. Consequently, the RGLE is basically parameter free, while the CGLE contains two relevant parameters \( c_1, c_3 \) that absorb all control parameter of the real
model. The behavior near a Turing bifurcation is thus universal and completely understood, while near a Hopf bifurcation a large variety of dynamical phenomena can appear depending on the values of the actual parameters $c_1$ and $c_3$. Spatiotemporal chaos in the one-dimensional CGLE will be investigated in Chapter 3 of this work. The third important generic case is the wave instability with $\Omega_C \neq 0$ and $k_C \neq 0$. The behavior near onset is then described by coupled CGLEs for the left and right traveling waves [45]. Further extensions arise when two instabilities appear simultaneously in a so-called codimension-2 bifurcation. Then the corresponding amplitude equations have to be coupled, for a review on the Turing-Hopf instability, see [46]. A study of amplitude equations valid near a Turing-Wave instability is presented in the next chapter. Studying amplitude equations has proved to be a useful supplement in the investigation of realistic reaction-diffusion systems [47]. It allows for easy comparisons with pattern formation in other fields such as hydrodynamics and nonlinear optics where amplitude equations are more common than in chemical and biological systems.

1.3.2 Numerical Stability Analysis of Traveling Waves

To analyze traveling waves in the analogous one-dimensional model, we switch to a reference frame moving with speed $c$. Replacing $x$ by $z = x - ct$ transforms the Eqs. (1.6) to:

$$\partial_t U(z, t) = F(U(z, t), \mu) + D \partial_z^2 U(z, t) - c \partial_z U(z, t) \quad (1.11)$$

Travelling waves $U_W(z) = U_W(x - ct)$ with speed $c$ will appear as nonuniform, steady solutions that fulfill the ordinary differential equation $F(U_W) + Dd_{zz}U_W - cd_zU_W = 0$. It is instructive to write the previous conditions for each component $U_i$ of $\vec{U}$ with $i = 1, \ldots, n$ and

$$F_i(\vec{U}(z)) + D_i \frac{d^2 U_i(z)}{dz^2} + c \partial_z \frac{dU_i(z)}{dz} = 0 \quad (1.12)$$

This can be easily transformed into two first order equations (ODEs) by introducing new variables $V_i$ if $D_i \neq 0$:

$$\frac{dU_i(z)}{dz} = V_i(z)$$

$$\frac{dV_i(z)}{dz} = -\frac{1}{D_i} \left(cV_i(z) + F_i(\vec{U}(z), \vec{P})\right) \quad (1.13)$$
or into a single first order equation if $D_i = 0$

$$\frac{dU_i(z)}{dz} = -\frac{1}{c} \left( F_i(\bar{U}(z), \bar{P}) \right). \quad (1.14)$$

Fixed points in these travelling wave ODEs correspond to spatially homogeneous steady states; limit cycles correspond to wave trains, fronts to heteroclinic orbits and pulses to homoclinic orbits, for illustration see Fig. 1.7.

Once traveling wave solutions are found, their linear stability can be deduced from the time evolution of small perturbation $u(z, t) = U(z, t) - U_W(z)$. Plugged into the
original PDE Eqs. (1.11), one obtains
\[ \partial_t u = \mathcal{L}(U_W(z), \mu)u + \mathcal{O}(u^2), \]  
(1.15)
where \( \mathcal{L} \) is the Jacobian with \( \mathcal{L}_{ij} = \partial_{U_j} F_i(U_W(z)) + D \partial_{zz} - c \partial_z \).

The problem consists of finding the eigenmodes of the linearization around a traveling wave solution in the appropriate functional space. The appropriate ansatz is
\[ u(z, t) = \sum_{n} \Phi_n(z) \exp(\omega_n t). \]  
(1.16)
This generates the eigenvalue problem
\[ \mathcal{L}(U_W(z), \bar{\mu})\Phi_n(z) = \omega_n \Phi_n(z) \]  
(1.17)
which decides if the solutions are stable (all \( Re\omega_n \leq 0 \)) or unstable (at least one eigenvalue with \( Re\omega_n > 0 \)). The eigenvalue problem maybe solvable in closed form, only if the traveling wave solution \( U_W \) is known in closed form. Since this is not the case for most models studied here, a different strategy is employed: the PDE is discretized in a large but finite domain mainly using dealiased pseudospectral (but also finite difference) methods. In subsequent chapters, such a stability analysis is applied to solitary pulses and periodic pulse \textit{resp.} wave trains. For pulses, periodic boundary conditions are used and the infinite system is approximated by a ring of finite length. While discretization introduces an error in the representation of large wavenumber modes, the finite ring length is a more critical parameter - in general one has to empirically find out which ring length is sufficient. Similar considerations hold for the stability analysis of rotating spiral waves, that is best carried out in corotating frame and spirals with a symmetry center in the middle of a circular domain (see Chapter 2).

For periodic traveling waves with a wavelength \( \lambda \), we can take advantage of Bloch’s theorem and fix the ring length to be \( L = \lambda \). Bloch’s theorem states [48] that the eigenfunctions \( \Phi \) are of the form
\[ \Phi_n(z) = e^{iqz} \phi_{nq}(z) \quad \text{with} \quad \phi_{nq}(z) = \phi_{nq}(z + \lambda), \]  
(1.18)
where for a train of \( n \) pulses in a ring of length \( L = n\lambda \), the wavenumber \( q \) fulfills \( q = m2\pi/L \) with \( m = 0, \ldots, n - 1 \) integer. Plugging this equation into Eq. (1.17) yields
\[ \mathcal{L}(U_W(z), q, \bar{\mu})\phi_{nq}(z) = \omega_{nq} \phi_{nq}(z). \]  
(1.19)
The operator $\mathcal{L}$ explicitly depends on the wavenumber $q$, where the possible $q$ are determined by the number of pulses $n$ that fit in the ring. In practice this means, that for doing the numerical stability of $n$-pulses on a ring of length $L = n\lambda$, one needs to do $n$ computations with a single pulse on a ring of length $L = \lambda$.

The procedure works well for one-dimensional reaction-diffusion systems, where the discretization typically produces $100 - 1000$ equations and the corresponding Jacobian can be diagonalized with standard procedures. In two dimensions, one has instead $10^4$ to $10^6$ ordinary differential equations and the corresponding Jacobians have thus $10^8 - 10^{12}$ elements. These Jacobians can no longer be diagonalized completely, instead iterative methods are used to obtain the largest eigenvalues [49]. Here, we constrain ourselves almost always to one-dimensional problems, only in Chapter 2 the problem of rotating spiral waves in small circular domains is treated. Numerical stability analysis proved to be useful for a great variety of phenomena including pulse interactions, interaction of spirals with reflecting walls (Chapter 2) as well as instability of pulses, wavetrains and rotating spirals (Chapter 3). It even allows insight into transitions between different phases of spatiotemporal chaos (Chapter 3).

### 1.3.3 Other Methods

The methods described in the previous subsection are the most relevant tools for the studies in this thesis. However, there is a variety of other useful methods that are listed briefly in the following. If we have localized structures such as fronts or pulses, we can often derive a reduced dynamics that assigns only a few degrees of freedom to a single front or pulse. This approach is fruitful for the theory of interacting pulses (Chapter 2) as well as for propagation of fronts in anisotropic and heterogeneous media (Chapter 4) and will be discussed in the respective parts of this work.

A cornerstone of all work in pattern formation is direct numerical integration of the nonlinear partial differential equations. For reaction-diffusion systems this task is less involved than for other application, e. g. hydrodynamic flows. We have used mostly finite-difference schemes with explicit, implicit or semi-implicit timesteppers. In some cases, for example, models with nonlocal coupling or the complex Ginzburg-Landau equations, pseudospectral methods are more efficient and have been employed.

In the last chapter (Chapter 5), we employed also some tools from the theory of stochastic processes as well as simulations with discrete models related to cellular
automata or models for surface growth.
2 Complex Patterns due to Competition and Interaction

This section deals with complex patterns arising from simultaneous presence of simpler patterns. In the first part, we analyze the dynamics for a medium with simultaneous appearance of traveling and stationary periodic patterns with different wave numbers. This bistability between different patterns leads to propagating interfaces and domains. It is reported that domains of constant size are structurally unstable near the onset of pattern formation. However, they are stabilized by the phenomenon of interface locking. In the remainder of the section, we are concerned with the interaction of simple patterns, namely pulses and spirals in excitable media. In the second section, we develop tools to study the interaction of pulses via their exponentially decaying tails. With a combination of simulations, kinematic theory, numerical bifurcation and stability results the problem of bound state formation and interaction of neighboring pulses is solved in a consistent way. The method is applied to a simple two-component activator-inhibitor model as well as to a realistic three-component model of a catalytic surface reaction.

Finally, we turn to the treatment of interaction of rotating spirals in reaction-diffusion media. Experimental and theoretical studies on the problem of interaction of a spiral with a mirror wall realized by a no-flux boundary condition are described. For reasons of symmetry, these studies have been conducted in small circular domains. Upon decrease of the domain size, the rotation frequency of spiral resp. rotating waves increases indicating a repulsive interaction of the spiral with the wall.
2.1 Competition between Traveling Waves and Turing Patterns

Pattern forming processes in nonequilibrium systems are associated with instabilities of the spatially homogeneous state. Cross and Hohenberg distinguish three basic types of instabilities in unbounded systems in their seminal review article on pattern formation [1]: (I) spatially periodic and stationary in time, (II) spatially periodic and oscillatory in time and (III) spatially homogeneous and oscillatory in time. In reaction-diffusion systems, these instabilities are usually called Turing, wave and Hopf bifurcation, respectively. Standard two variable reaction-diffusion equations of activator-inhibitor type allow only for the Turing and Hopf instability. A particular interesting situation appears when Turing and Hopf modes appear simultaneously at a so called Turing-Hopf point [46]. The normal form is then simply given by a coupling of the two respective amplitude equations. To obtain a wave instability, we need to include a third variable. This is done here; we will consider a particular limit of this three variable model, that leads to a two-variable model with nonlocal coupling. The model also allows for Turing instabilities. They compete with the wave instabilities and coincide with them at the Turing-wave point. We derive the normal form for this point and study the case of competition between stable Turing patterns and waves.

We start from the three variable system

\[\begin{align*}
\partial_t u &= au + \beta u^2 - \alpha u^3 - bv - gw + \partial_x^2 u \\
\partial_t v &= cu - dv + \delta \partial_x^2 v \\
\tau_w \partial_t w &= eu - fw + \gamma \partial_x^2 w
\end{align*}\] (2.1)

These equations are an extension of the FitzHugh-Nagumo model by a second inhibitor \(w\). In the limit of a fast second inhibitor \(\tau_w = 0\), one finds

\[\begin{align*}
\partial_t u &= au + \beta u^2 - \alpha u^3 - bv + \partial_x^2 u \\
&\quad - \mu \int_{-\infty}^{+\infty} e^{-|x-x'|} |u(x',t)| dx' \\
\partial_t v &= cu - dv + \delta \partial_x^2 v.
\end{align*}\] (2.2)

The nonlocal coupling in the \(u\)-dynamics arises from the adiabatic elimination of the fast inhibitor \(w\) in Eqs. (2.1). For analytical investigations, we consider the model on the infinite line, while numerical simulations are typically performed in large systems
Bistability between steady states

Traveling fronts and pulses

Bistability between pattern families

Interfaces

Drifting pattern domains

Fig. 2.1: Schematic construction of drifting pattern domains
with periodic boundaries. The parameters characterizing the inhibitory nonlocal coupling in Eqs. (2.2) are then found as \( \sigma = \sqrt{f/\gamma} \) and \( \mu = g\sqrt{e^2/\gamma f} \) from the original three variable model above. The emphasis here is on the onset of pattern formation resulting from destabilization of a single homogeneous steady state. Eqs. (2.2) possess the trivial homogeneous fixed point \( \mathbf{u}_0 = (u_0, v_0)^T = (0, 0)^T \) for all parameter values. Here, we consider the regime where this fixed point is the only one present, i.e. \( a < bc/d + 2\mu/\sigma \) and consider perturbations \( e^{ikx-\lambda(k)t} \), where \( \lambda(k) = \chi(k) + i\omega(k) \). The growth rates \( \lambda(k) \) are given by the eigenvalues of the Jacobian. Linear stability analysis reveals that Eqs. (2.2) exhibit wave instabilities if the nonlocal coupling is of sufficiently long range \( \sigma < \sigma_c = (2\mu/(1 + \delta))^{1/3} \).

In the following we vary the control parameters \( a \) and \( \delta \); the „driving force” \( a \) represents the kinetics, whereas the ratio of diffusion coefficients \( \delta \) describes the spatial coupling in the medium. All other parameters of Eqs. (2.2) have been fixed. For the wave bifurcation, the critical wavenumber \( k^c_W \) and parameters \( a_W, \delta_W \) are obtained from the condition \( \lambda(k^c_W) = \pm i\omega_0 \) where the perturbation with \( k^c_W \) is the fastest growing mode with \( (k^c_W)^2 = \sqrt{2\mu/1+\delta} - \sigma^2 \). Note, that for both \( \sigma = \sigma_c \) and \( \sigma = 0 \) (global coupling limit) the critical wavenumber is \( k^c_W = 0 \). Similarly, a competing Turing instability appears for a critical parameter \( a_T \) with a wavenumber \( k^c_T \), where the leading eigenvalue \( \lambda(k^c_T) = 0 \). For large enough driving \( a \), the wave instability appears for small \( \delta \), while for large \( \delta \) the Turing instability destabilizes the homogeneous state. For the chosen parameter values, the system exhibits a Turing-wave point (TWB), for which a stationary Turing mode and two oscillatory, traveling wave modes become unstable simultaneously.

Near the TWB, we can derive a set of coupled equations for the amplitudes \( A, B \) and \( \mathcal{R} \) for left- and right-going waves and the Turing pattern, respectively. The amplitudes \( A, B \) and \( \mathcal{R} \) depend only on slow time and space variables. The resulting equations read

\[
\begin{align*}
\partial_t \mathcal{R} &= \eta \mathcal{R} - |\mathcal{R}|^2 \mathcal{R} + \xi \partial_x^2 \mathcal{R} - \zeta (|A|^2 + |B|^2) \mathcal{R} \\
\partial_t A + c_g \partial_x A &= \rho A + (1 + ic_1) \partial_x^2 A - (1 - ic_3)|A|^2 A \\
&\quad - g(1 - ic_2)|B|^2 A - \nu(1 - i\kappa)|\mathcal{R}|^2 A \\
\partial_t B - c_g \partial_x B &= \rho B + (1 + ic_1) \partial_x^2 B - (1 - ic_3)|B|^2 B \\
&\quad - g(1 - ic_2)|A|^2 B - \nu(1 - i\kappa)|\mathcal{R}|^2 B.
\end{align*}
\]

Note, that the nonlocal term of Eqs. (2.2) only enters into the diffusion coefficients of Eqs. (2.3) and does not give rise to a nonlocal term in Eqs. (2.3). Knowledge of the
coefficients of Eqs. (2.3) allows analytical predictions of the pattern dynamics. Here, traveling waves are always preferred over standing waves ($g > 1$, see [1]) and bistability between wave and Turing patterns is found ($\nu \zeta > 1$). In this bistability region in parameter space, a family of stable Turing patterns and two families of stable left- and right-traveling waves parametrized by their corresponding wavenumbers coexist.

The bistability between Turing and wave patterns poses an interesting question. It is well known, that fronts [50] and pulses [51] appear in bistable media with coexisting stable homogeneous steady states. Here, we have bistability between two families of periodic patterns, namely stationary Turing structures and traveling waves. The above model allows us to study structures that are analogue to fronts and pulses in these media, namely interfaces between Turing and wave patterns, see sketch in Fig. 2.1. A new aspect is that we have now not two unique coexisting states, but instead two families of periodic patterns (stationary and traveling) that are parametrized by their respective wavenumbers and frequencies. This poses the question of wavenumber selection on both sides of the interface similar to previous studies of sinks and sources in systems with wave bifurcations [45]. A second related issue is the possibility that a stable domain of one pattern is embedded into a background of the other pattern, see Fig. 2.2, so called drifting pattern domains.
Drifting pattern domains are obtained in a subregion of the bistability range between Turing and wave patterns in simulations of the original reaction-diffusion model, Eqs. (2.2). Starting with a traveling wave pattern at slow inhibitor diffusion ($\delta < 1$, Fig. 2.2a), we increase the parameter $\delta$ towards the regime of Turing patterns ($\delta > 1$, Fig. 2.2b). In some cases, a sudden transition happens along the way, in others intermediate mixed states with drifting pattern domains appear (see Fig. 2.2c,d). They may contain only a single element of one of the two patterns (see Fig. 2.2e,f). Both issues are discussed in more depth in [52] and in great detail in [53]. Drifting pattern domains (DPDs) of fixed width are found to be created by a „non-adiabatic“ locking effect of interfaces between Turing and wave patterns in Eqs. (2.2). Consequently, DPDs of constant width are not structurally stable in the amplitude equations (2.3) where the fast spatial and temporal variation have been adiabatically eliminated.

While DPDs have not been reported in experimental reaction-diffusion systems, strikingly similarly patterns have recently been found in a hydrodynamic system - arrays of liquid columns [54]. These patterns are shown in Fig. 2.3. DPDs can be excited from the initially stationary liquid columns by a local perturbation of the wavelength (see circle in the upper right corner of Fig. 2.3a). They maintain constant width (Fig. 2.3b). A global perturbation gives rise to traveling columns (Fig. 2.3c), inside which again small drifting domains of stationary patterns may form (Fig. 2.3d).
2.2 Interaction of Pulses in Excitable Media

Propagating pulses with constant speed can be generated by locally perturbing an excitable medium sufficiently strong. In chemical systems, such excitations correspond to concentration waves and are often observed in catalytic reactions in solutions or on surfaces. In biology, important examples are the propagation of electrical impulses in neurons and on the heart muscle. Here, coherent structures - solitary pulses resp. pulse trains - transmit information. The interactions between such coherent structures are investigated in this section. This study contributes to the understanding of patterns, which consist of ensembles of such coherent structures, e. g. periodic and irregular pulse trains.

Excitable processes are well modelled by reaction diffusion equations [55]. Their variables are identified with concentrations in chemical reactions or densities of cells in aggregation processes. Often these pattern-forming systems are of activator-inhibitor type, where spatiotemporal dynamics result from the interaction between a fast autocatalytic and diffusing substance, the activator, and a slow reacting inhibitor species. The stable rest state then corresponds to low values of activator and inhibitor concentrations. A local rise of activator concentration leads to excitation, a strong amplification and rapid growth of the activator concentration towards the excited state. Inhibitor production follows slowly in the excited state. If the inhibitor, however, reaches a critical level, a rapid decay of the high activator concentration back to the low values near the rest state is observed. The subsequent removal of the remaining inhibitor characterizes the refractory state.

The interaction of pulses and the resulting pulse dynamics were investigated in great detail for the case of a simple two-variable activator-inhibitor model for a chemical reaction in a one-dimensional ring (periodic boundary conditions)

\[
\begin{align*}
\partial_t u &= \frac{1}{\epsilon} u(u-1)(u-a) + \partial_x^2 u, \\
\partial_t v &= h(u) - v,
\end{align*}
\]

where \(h(u)\) is a cubic function, see also [56]. The parameters are chosen to realize an excitable medium of type II. The medium persists in the rest state, which is stable against small perturbations, until a pulse is excited. After passage of this pulse, a recovery time ("refractory phase") follows, in which the medium cannot be easily excited again. Because of this refractory phase, a pulse following another pulse propagates slower than its predecessor. The velocity of a pulse decreases with the distance
Complex Patterns due to Competition and Interaction

Fig. 2.4: Experimental example of pulse interaction in the NO + CO reaction

to its predecessor. Consequently, the velocity of a wavetrain often decreases with the wavelength (normal dispersion). In fact, the standard choice of linear inhibitor production with \( h(u) = u \) used in the FitzHugh-Nagumo equations in the previous chapter allows only for normal dispersion. Different forms of dispersion are found, when \( h(u) \) is nonlinear, e. g. a cubic function, see [56].

In new experiments concerning the catalytic reaction of CO and NO on a Pt-surface [57], an interesting behaviour was found: Pulses get faster with decreasing distance (anomalous dispersion), see Fig. 2.4. As a consequence of this type of interaction, pulses can merge, annihilate each other or they can form stable bound pairs. Note, that for a pair of pulses, anomalous (normal) dispersion corresponds to an attractive (repulsive) interaction. The above experiments can be reproduced in numerical simulations with a mechanistic model of the NO + CO reaction [58, 59] based on the reaction scheme shown in Fig. 2.5. The resulting equations read in the one-dimensional version

\[
\partial_t u = k_1 p_{CO}(1 - u - v) - k_2(u, v)u - k_3uw + D\partial_x^2 u
\]
Fig. 2.5: Scheme for catalytic reduction of NO with CO on a Pt(100) surface.

\[
\begin{align*}
\partial_t v &= k_1 p_{NO}(1 - u - v) - k_4(u, v)v - k_5 v f(u + v, w) + D\partial^2_x v \\
\partial_t w &= k_5 v f(u + v, w) - k_3 uw
\end{align*}
\] (2.5)

and describes the spatiotemporal evolution of the surface concentrations of CO \(u\), NO \(v\) and oxygen \(w\). For simplicity, the diffusion constants of CO and NO are set equal: \(D_{CO} = D_{NO} = D\). More details on the experiments and simulation results are documented in [57].

To study interactions of pulses on a ring, equations for reduced dynamics reduced dynamics can be established. Here, only the influence of the interactions on the velocity of each involved pulse is considered. The interactions are determined by numerically computing the dispersion curve \(c(d)\). The reduced dynamical equations can be derived asymptotically from perturbation theory assuming large interpulse distances [60]. Alternatively, one can use kinematical theory [61] based on the knowledge of the dispersion curve \(c(d)\). Equations for the positions \(p_1, p_2 > p_1\) of a pair pulses read

\[
\begin{align*}
\dot{p}_1 &= c(d_1), \\
\dot{p}_2 &= c(d_2) = c(L - d_1),
\end{align*}
\] (2.6)

where \(d_1 = p_2 - p_1\). In the case of a pair of pulses moving with equal speed and distances \(d_1, d_2\) on a ring, we transform the equations to a co-moving frame. Therein, a family of steady state solutions \(p_1^0, p_2^0\) exists. Linearization of the reduced dynamics then yields the following evolution equations for perturbations \(\delta p_i\) of the steady state
pulse positions $p_1, p_2$:
\[
\begin{align*}
\delta p_1 &= c'(d_1) \cdot (\delta p_2 - \delta p_1), \\
\delta p_2 &= c'(d_2) \cdot (\delta p_1 - \delta p_2).
\end{align*}
\] (2.7)

This problem leads to the eigenvalues
\[
\lambda_1 = 0 \quad \text{and} \quad \lambda_2 = -(c'(d_1) + c'(d_2))
\] (2.8)

with the respective eigenvectors
\[
\vec{e}_1 = \begin{pmatrix} 1 \\ 1 \end{pmatrix} \quad \text{and} \quad \vec{e}_2 = \begin{pmatrix} -\frac{c'(d_1)}{c'(d_2)} \\ 1 \end{pmatrix}.
\] (2.9)

The eigenvector $\vec{e}_1$ corresponds to a shift of both pulses in the same direction. Due to the translational invariance of the system, the eigenvalue $\lambda_1 = 0$. The eigenvector $\vec{e}_2$ corresponds to a relative shift of pulse positions and thus yields a change in the inter-pulse distance. Thus, it is termed the “interaction” eigenvector with the “interaction” eigenvalue $\lambda_2$. The sign of $\lambda_2$ is given by the sum of the slopes of the dispersion curve at $d_1$ and $d_2$. For an equidistant pulse pair ($d_1 = d_2$), a negative (positive) slope yields instability (stability) against such a shift. A new branch of solutions with $d_1 \neq d_2$ (non-equidistant pair) can be easily obtained from the reduced pulse dynamics, if the dispersion curve is nonmonotonous. Then, a given speed $c$ may correspond to two or more distances $d_i$. To check the quality of the reduced equations, we computed interaction eigenvalues and eigenfunctions explicitly by numerical stability analysis.

Fig. 2.6 shows equidistant and nonequidistant pairs of traveling pulses in Eqs. (1.4) in the case of anomalous dispersion. The equidistant pulse pair (dashed red line in Fig. 2.6A) is unstable and is destroyed by pulse attraction. The new stable solution is given by the non-equidistant pulse pair (solid black line in Fig. 2.6A). Fig. 2.6B shows the $u-$components of the leading eigenmodes, where the red dashed curve represents the only mode with a positive eigenvalue $\lambda_2$ in the equidistant pair. Its antisymmetric shape corresponds to the eigenvector $\vec{e}_2$ of the reduced dynamics. The eigenfunction corresponding to $\vec{e}_2$ of the non-equidistant pulse pair (solid black curve in Fig. 2.6B), on the other hand, has a negative eigenvalue and thus stabilizes the pulse pair. The destabilization near the maximum of the dispersion curve (red curve in Fig.2.6C), and the bifurcation of a non-equidistant pulse pair have been confirmed. The new branch is shown as solid black line. Fig. 2.7 shows a comparison of the eigenvalues gained from reduced dynamics and the leading eigenvalues from numerical stability analysis.
While the results show reasonable agreement in the case of the equidistant pulse pair, the prediction for the nonequidistant pairs are less satisfying. Thus, numerical stability analysis is essential to compute the stability of bound pairs.

The dispersion curve (red curve in Fig. 2.6C ) shows a transition from normal dispersion with a positive slope (red solid line) to anomalous dispersion with a negative slope (red dashed line). In the range of anomalous dispersion, a second branch of stable solutions corresponding to non-equidistant pulse configurations exists (black curve in Fig. 2.6C). In an infinitely long system, this branch represents a bound pair of pulses. Thus, bound pairs can be traced back to a pitchfork bifurcation near an extremum of the dispersion curve at finite wavelength. With the same methods bound states of three or more pulses have been calculated and their stability in dependence of the system parameters has been determined, see [56].

Calculations with the realistic model of the NO and CO reaction provide a qualitatively similar dispersion curve. There, pulses do not form bound states. Instead one of the pulses disappears as soon as it gets too close to the other one in agreement with laboratory experiments [57]. The described theory of pulse interactions has been recently applied also to the realistic model of the NO reduction with CO [62]. The main new result concerns the observed merging of the two pulses. While bound pairs still exist, they are destabilized by an oscillatory internal mode above a critical length of the ring. Finally, it is interesting to mention that anomalous dispersion has recently discovered in a variant of the Belousov-Zhabotinsky reaction [63]. The authors of the latter study report beside the merging of pulses also the formation of bound states of many pulses („pulse bunching“) in line with our theory of pulse interactions presented here and in [56].
Fig. 2.6: (A): Pair of pulses on a ring of length $L$. The solution with the pulse distance $L/2$ is unstable (red line). The black line shows a bound pair with a distance considerably smaller than $L/2$. (B): The interaction eigenmodes corresponding to the solutions in diagram (A). The black mode belongs to a negative and the red one to a positive eigenvalue. (C): The red curve corresponds to a branch of equidistant pulse pairs in a ring with length $L$, where the solid (dashed) line symbolizes stability (instability). It gives also the dispersion curve for pulse trains with pulse distance $L/2$. The black curve represents stable, non-equidistant pulse pair solutions.
Fig. 2.7: Comparison of the eigenvalues (4) of the reduced dynamics (blue and red lines) with results of numerical stability analysis (circles) for the case of equidistant (A) and non-equidistant (B) uniformly moving pulse pairs.
2.3 Interaction of Spiral Waves

**Note:** The results presented in this section have not been previously published and are not contained in the appendix of the present thesis. They have been obtained in collaboration with I. G. Kevrekidis (Princeton). Therefore we choose to present the results in more detail than in other parts of this thesis.

2.3.1 Introduction

Rotating spiral waves constitute one of the most common spatiotemporal patterns observed in two dimensional excitable media far from equilibrium conditions. They have been observed in a wide variety of systems including heterogeneous catalytic reactions \((CO + O_2/Pt, NO + CO/Pt \text{ etc.})\), liquid phase reactions (such as the famous Belousov-Zhabotinskii \((BZ)\) reaction), slime mold aggregation, and electrical activity on cardiac tissue. Their ubiquitous presence has generated a widespread interest in the study of their dynamical behavior among physicists, biologists and applied mathematicians \([17, 64, 65, 66, 67, 68]\).

The experimental investigations of spiral waves in earlier years almost exclusively concentrated on the \(BZ\) system \([69, 70]\). The reaction can be carried out easily in a petri-dish (batch) or using a continuous setup. More recently, a lot of interesting observations have been made on the CO oxidation system \([71]\). One motivation for the study of spirals comes from cardiology; the breakup of isolated spiral waves of electrical activity on the cardiac tissue into disorganized excitations is deemed to play an important role in fibrillation \([65]\). Spiral interaction has been extensively studied in the complex Ginzburg-Landau equation (CGLE) by various authors. Controversial ideas about the range of the interaction force have been published, while early work favored a \(1/r\) decay \([78, 79]\) more extensive recent analysis and simulations point towards an exponential decay \([80]\). Another important issue deals with formation of symmetric spiral pairs or spontaneous symmetry breaking and decay of one of two spirals in an interaction event. In reaction-diffusion models for excitable media, However a related problem has been studied with quite some effort and will be discussed in some detail below: the interaction of spirals with a reflecting mirror wall in a circular geometry. Kinematical theory predicts a \(1/r\) correction to the rotation frequency \([81]\), while a different approach yields a superexponential correction \([82]\). Experiments on a catalytic surface in small circular domains and corresponding simulation are documented.
They show a particular strong effect of the domain size for weak excitability where the kinematic theory is valid. Simulations in this regime show a $1/r$-correction, see [83], as predicted by kinematical arguments [81]. The superexponential effects in the limit of good excitability [82] predicts only extremely weak changes in the rotation frequency. All theoretical results are based on some oversimplification of realistic models. Numerical simulation can give an approximate picture, but more accurate results can be obtained by means of numerical stability analysis of spirals as shown in the following. Before we enter into the topic of numerical stability analysis of spirals in small domains, we review briefly the general properties of rotating spirals.

2.3.2 Dynamics of Rotating Spirals

Most theoretical studies on the spiral waves have centered around two limiting cases. The free boundary approach was first introduced by Tyson and Keener [66] and later investigated by others [65, 67, 72, 73]. A simple kinematic theory in terms of motion of curves was first sketched in 1946 by Wiener and Rosenblueth [19] and later carried out by Zykov et. al. [64, 74]. Although the analytical work has contributed to a significant advancement in the understanding of spiral wave behavior, it has proven to be limited and difficult. Numerical investigations of the dynamics of spiral waves have been carried out for a variety of reaction-diffusion systems [25, 75, 76, 77].

It is worthwhile to look at the two-parameter phase diagram of wave dynamics in a generic model of excitable media shown in Fig. 2.8, reproduced from an article by Winfree [17]. The phase diagram is „generic” in the sense that most two-parameter investigations of spiral dynamics in other excitable systems have shown similar states and transitions. This diagram was obtained by Winfree in a detailed numerical study of the Fitzhugh-Nagumo (FHN) equations introduced in Chapter 1, which read

\begin{align}
\partial_t u &= \nabla^2 u + u - \frac{u^3}{3} - v, \\
\partial_t v &= \epsilon (u + \beta - \gamma v).
\end{align}

The parameter $\gamma$ was fixed at 0.5 and numerical simulations were conducted to observe the various states shown in the figure.

The two-parameter phase diagram in $(\epsilon, \beta)$ is divided into five distinct regions representing different dynamical states, separated by curves of bifurcation loci. When $\beta$ and $\epsilon$ are large (in the lower left end of the figure), no wave propagation is possible.
Fig. 2.8: Two parameter phase diagram of the spiral wave dynamics for the FHN model with $\gamma = 0.5$, reproduced from [17]. $\partial P$ denotes the propagation boundary; $\partial R$ denotes the rotor boundary; $\partial M$ and $\partial C$ denote transitions to quasiperiodic and complex states respectively. Also shown are tip paths centered at corresponding parameter points. Circles correspond to rotating waves and “flowers” to meandering and complex states.
in the FHN system. Here the medium is not sufficiently excitable to support waves and all initial conditions evolve to a spatially uniform state. The curve $\partial P$ denotes the “boundary of propagation”; planar waves exist in the system above this curve. The curve $\partial R$ denotes the “rotor boundary”; the system supports rotating wave solutions above this curve. A good representation of the dynamics near the $\partial R$ boundary is the evolution of a broken wave segment of a planar wave [64]. In the region between $\partial P$ and $\partial R$, broken wave tips retract as they propagate along straight lines. Above the $\partial R$ curve, an initial broken segment curls up and evolves into a rotating spiral wave. Spirals exist in the region bounded by $\partial R$ and $\partial M$ curves, and are characterized by rigid rotation around a fixed point. The tip paths of the waves are also superimposed on the Fig. 2.8; each tip trace is centered on the corresponding parameter point. Tip motions of rigidly rotating spirals form circles. The radius of the tip path, spiral period and spiral wavelength all diverge as the boundary $\partial R$ is approached in the parameter space. The curve $\partial M$ denotes the “meandering boundary” and is associated with the transition from rigidly rotating spirals to meandering motion of the spiral core. Using a similar model of excitable media, Barkley [68] showed that $\partial M$ is a single smooth locus of Hopf bifurcation of rotating spirals. The tip of the meandering spirals (more specifically modulated rotating waves) form flower-like patterns consisting of two frequencies (see the region between $\partial M$ and $\partial C$ in the two phase diagram). Finally, the region to the right of the $\partial C$ curve is characterized by dynamics that are more complicated than two-frequency quasiperiodic, and possibly even chaotic. These states are termed by Winfree as “hyper-meandering”.

Although accurate numerical simulations provide useful tools for analysis, there are still many open problems that can be tackled better by a detailed linear stability and bifurcation analysis. In this chapter, we will present an outline of the computational tools that we have developed for bifurcation studies of rotating spirals waves. Some results on the instabilities of spirals in small circular domains are then presented.

### 2.3.3 Linear Stability Analysis of Spirals

Spiral waves are solutions to the governing reaction-diffusion equations with a rotational symmetry; they appear stationary in a frame rotating with the frequency of the wave. Consequently, they can be computed as steady states of the following reaction-diffusion equations in polar coordinates:

$$0 = \partial_t u = F(u) \equiv f(u) + D \nabla^2 u + \omega \partial_\theta u$$  \hspace{1cm} (2.12)
where \( \mathbf{u} = (u, v)^T \), \( \mathbf{D} = \text{diag}(1, 0) \), and \( \mathbf{f}(\mathbf{u}) \) represents the kinetic terms of a modified Barkley model with delayed inhibitor production

\[
\mathbf{f}(\mathbf{u}) = (-\epsilon^{-1}u(u - 1)(u - (v + b)/a), u^3 - v)^T.
\]

This model is a simplified variant of the one used in the section on pulse interactions.

The last term on the right hand side in Eq. (2.12) comes from the rotating frame of reference. The boundary conditions on a circular domain of radius \( R \) are taken to be no flux in the radial direction \( (\partial_r \mathbf{u})|_{R} = 0 \). The system defined by Eq. (2.12) has a continuum of solutions (given by all arbitrary rotations of the spiral) and an additional parameter \( \omega \). This indeterminacy is removed and a unique steady solution is picked out by fixing the rotational phase of the spiral at \( (r = R/2, \theta = \pi) \). This is done by appending an additional pinning condition given by,

\[
\partial_{\theta} v \bigg|_{r = R/2, \theta = \pi} = 0, \tag{2.13}
\]

which allows for determining the unique value of \( \omega \).

The stability of the steady state \( \mathbf{u} \) of Eqs. (2.12) and (2.13) with respect to small perturbations is determined by the following linearized eigenvalue problem:

\[
\mathbf{D}(\mathbf{u})\mathbf{U} = \lambda \mathbf{U} \tag{2.14}
\]

where

\[
\mathbf{D}(\mathbf{u}) = \mathbf{D}(\mathbf{u}) + \mathbf{D} \nabla^2 + \omega \partial_{\theta} \tag{2.15}
\]

while \( \lambda \) and \( \mathbf{U} \) are the eigenvalues and the eigenmodes of the linearized operator \( \mathbf{D}(\mathbf{u}) \).

The eigenvalues \( \lambda \) determine the linear stability of the spiral solution; a bifurcation is indicated when a real eigenvalue or an imaginary pair cross into the right half complex plane.

Eqs. (2.12) are discretized on a polar grid by expanding the concentration fields \( u \) and \( v \) in Fourier modes in the azimuthal direction and using second order finite differences to evaluate the operators in the radial direction,

\[
\nabla_\theta^2 f(r, \theta) = \frac{1}{r^2} \frac{\partial^2}{\partial \theta^2} f(r, \theta) \rightarrow -\frac{k^2}{r^2} f_k(r, \theta)
\]

\[
\nabla_r^2 f(r, \theta) = \frac{1}{r} \frac{\partial}{\partial r} f(r, \theta) + \frac{\partial^2}{\partial r^2} f(r, \theta) \rightarrow \frac{f_{i+1} - f_{i-1}}{2r \Delta r} + \frac{f_{i+1} + f_{i-1} - 2f_i}{\Delta r^2},
\]

\( \Delta r \) is the radial grid spacing.
where the $f_i$ are the values of the field $f$ on which the above operators act at discrete sites of a polar grid. $f_{i-1}, f_{i+1}$ are the values at next neighboring sites in the radial direction. The boundary conditions at $r = R$ are taken to be no-flux in the radial direction by setting,

$$\nabla^2_r |_{r=R} = \frac{\partial^2}{\partial r^2} |_{r=R} = 2 \frac{f_{N-1} - f_N}{\Delta r^2}.$$  

The boundary condition at $r = 0$ is taken to be zero-flux as well, while the singularity in the first term in Eq. (2.16) is treated by using L'Hôpital’s rule,

$$\nabla^2_r |_{r=0} = \lim_{r \to 0} \frac{\partial}{r \partial r} + \frac{\partial^2}{\partial r^2} = \frac{\partial^2}{\partial r^2} + \frac{\partial^2}{\partial r^2} = 4 \frac{\bar{f}_1 - f_0}{\Delta r^2}$$

where $\bar{f}_1$ is the averaged value of the field $f$ at $r = \Delta r$, while $f_0$ is the value of the field $f$ in the origin of the polar grid. Spatial discretization yields a set of ordinary differential equations describing the time evolution of coefficients of Fourier modes for the concentration variables. Most results reported here were obtained by taking $N_r = 51$ radial grid points and 30 Fourier modes in the azimuthal direction with $N_\theta = 128$ collocation points. Such a discretization yields a dynamical system of $\sim 6000$ ODEs. The geometric singularity in the mesh at $r = 0$ renders these ODEs very stiff and explicit time integrations methods cannot be utilized due to stability constraints. We perform transient simulations of the discretized system with the help of a stiff ODE solver called ODESSA [84], which employs variable (adaptive) step size with up to fifth order backward difference formulas.

Dynamical systems of this size are prohibitive for the routine stability and bifurcation algorithms based on direct solvers. We employ advanced iterative methods that have recently been developed and applied to large scale eigenproblems [49, 85, 86]. The steady state problem is solved using Newton’s method which can be accelerated by the use of Krylov-based iterative methods for linear systems [87]. Pseudoarclength continuation techniques are then utilized to follow the branch of spiral wave solution as the parameter values are changed. This continuation scheme is robust to the presence of folds in the spiral branch and traces the solution branch into parameter regions where the solution is unstable. The stability of the spiral solution is monitored by finding the leading (those with the largest real part) eigenvalues and corresponding eigenvectors at each continuation step. The leading eigenpairs are computed using an iterative Arnoldi method with implicit deflation [49, 85, 86].
Fig. 2.9: Eigenmodes due to symmetry: (a) $v$ concentration field of a rotating spiral wave; the parameters are $\epsilon = 0.025, a = 0.8, b = 0.02, R = 10$, (b) eigenmode resulting from rotational symmetry, (c) and (d) real and imaginary parts of the eigenmodes associated to the translational symmetry. The greyscale value represents the concentration of $v$; darker regions correspond to higher values of $v$ and vice versa.
Three of the eigenvalues of the linearized eigenproblem (2.14) lie on the imaginary axis and come from the symmetries in the infinite, unbounded system. Fig. 2.9 shows the spatial plots of the corresponding eigenmodes. The eigenvalue at zero \( \lambda_R \) comes from rotational symmetry of the spiral wave. The spatial contour plot of the corresponding eigenmode is displayed in Fig. 2.9. The rotationally symmetric eigenmode can be shown to be simply the azimuthal derivative of the concentration field, \( U_R = \partial_\theta \mathbf{u} \). Furthermore, a spiral wave in an infinite medium has a translational symmetry. In a frame rotating with the frequency \( \omega \), this results in a complex eigenpair at \( \lambda_T = \pm i\omega \) [68]. The real and complex part of the corresponding eigenmodes can be shown to be the \( x \) and \( y \) derivatives of the concentration field \( U_T = \partial_x \mathbf{u} \pm i \partial_y \mathbf{u} \).

The presence of no-flux boundaries in a finite domain at \( r = R \) breaks the translational symmetry of the spiral. However for a sufficiently large domain size, the eigenvalues associated with the translational symmetry are numerically very close to their values for an infinitely extended domain \( \lambda_T = 0 \pm i\omega \) [68]. The deviation of \( Re(\lambda_T) \) from zero is a measure of how much the boundaries influence the spiral core. Thus, the eigenvalues \( \lambda_T \) describe the interaction of the rotating spiral with the boundary. Since the spiral is centered in the circular domain, any translation will move it towards the boundary and should probe the force between the spiral and the zero-flux boundary, which can be considered a (curved) mirror wall. Consequently, the real parts of the eigenvalues \( \lambda_T \) of the rotating spiral in a corotating small circular domain correspond to the interaction eigenvalues \( \lambda_2 \) in the problem of pulse-pulse interaction, described in the previous section.

### 2.3.4 Spiral Instability in Small Circular Domains

Spirals in spatially extended domains select a unique set of values for their frequency \( \omega \) and spatial wavelength \( \lambda \) depending on the operating conditions. Recent work has shown that the presence of boundaries can significantly affect the behavior of waves in confined geometries. The presence of sharp corners may cause spiral nucleation from planar waves [88], while existing spirals are predicted to drift along the boundaries [89]. A number of experiments probe the dynamics of spirals near a boundary in the Belousov-Zhabotinsky reaction [90, 91] and find a measurable force that leads to meandering drift along the boundary. The effect of domain size on rotating waves has recently studied experimentally by Hartmann et. al., see [83] for the \( NO + CO \) reaction on a microstructured \( Pt(100) \) surface. The frequency of rotating waves was observed
to increase substantially for domain sizes below a critical domain size (that depends on spiral wavelength in extended domains) due to the interaction of the wave tip with the boundary. A linear increase in the spiral frequency with inverse domain size was analytically predicted by Davydov and Zykov [81] and supported by numerical simulations, see [83]. Furthermore, close to the onset of the frequency increase, a transition to quasiperiodic spirals was observed in simulations. After this transition, the spiral core is observed to drift near the boundary; we refer to this phenomenon as **boundary induced drift**.

We computed the stability of spirals in small circular domains using the model introduced in the previous section. Time evolution of a suitable initial condition was performed to obtain an initial spiral wave for a circular domain of size \( R = 5 \) units. Dimensionless parameters were taken in the typical excitable conditions as \( \epsilon = 0.025, a = 0.8, b = 0.02 \). The wavelength of a spiral in a large domain is about 9.1.

Starting from an initial guess of the spiral and its period, steady state continuation of the spiral solution was performed using the domain size \( R \) as the bifurcation parameter.

The bifurcation diagram of the spiral solution branch is displayed in Fig. 2.10(a) where the spiral period is plotted against the domain size. Fig. 2.10 (b) and (c) display the real and the imaginary parts of the eigenvalue that is associated with the translational symmetry of the spiral in the unbounded medium. When the domain size is above a critical value \( R_{cr,1} \), the spiral frequency remains essentially independent of the domain size. Note, that \( R_{cr,1} \) is less than half of the wavelength of the spiral in large domains in line with the observations in simulations of the standard Barkley model reported in [83]. Furthermore, the eigenvalues corresponding to the (broken) translational symmetry are close to its ideal value for a spiral in an infinite medium, \( \lambda_T \approx 0 \pm i \frac{2\pi}{T} \). Therefore, the value of \( R_{cr,1} \) gives a lower bound on the domain size above which the domain boundaries do not influence the spiral core.

For \( R < R_{cr,1} \) the spiral period is observed to decrease almost linearly with \( (R - R_{cr,1}) \). At the same time, the eigenvalue pair associated with the translational symmetry \( \lambda_T \) is seen to move into the left part of the complex plane. There exists another critical radius \( R_{cr,2} \) such that for domain sizes in between the two limits \( (R_{cr,2} < R < R_{cr,1}) \) the eigenvalue pair \( \lambda_T \) lies in the stable region \( (Re(\lambda_T) < 0) \). In this regime, spirals that are either perturbed away from the center or initiated off-center drift towards the center of the domain. At \( R = R_{cr,2} \), the eigenvalues associated with the translational
Fig. 2.10: Bifurcation diagram of a spiral wave in a circular domain. The period is plotted as a function of the circle radius $R$. The bottom two plots show the real and imaginary part of the eigenvalue associated with the translational symmetry.
symmetry cross out into the right half plane in a Hopf bifurcation. The stable solution after this bifurcation is a quasiperiodic spiral that drifts along the boundaries; therefore this transition marks the onset of “boundary induced drift”.

The quasiperiodic spiral solution exists up to a certain cutoff domain size $R_{cr,3}$ below which the medium does not support any rotating waves. The unstable spiral branch when continued further down in $R$ turns around in a saddle node bifurcation and then proceeds to collide with a spatially uniform steady state of the system at some critical domain size $R_{cr,4}$.

### 2.3.5 Transition to Meandering Spirals

We demonstrated in the previous section that the influence of the boundaries in smaller domains causes a quasiperiodic instability of the spiral core. The eigenmodes corresponding to this instability are the ones associated with the translational symmetry of the spirals in an unbounded domain. We refer to this instability as “boundary-induced drift” or “boundary-induced meandering”. Spirals in extended domains are known to exhibit another qualitatively different type of transition to quasiperiodic motion, which was illustrated for the FHN model in Fig. 2.8 at the $\partial M$ curve. Barkley [68] showed that this meandering transition is caused by a Hopf bifurcation of a set of isolated eigenvalues (that are different from the eigenvalues corresponding to the translational invariance). This instability will be referred to as “regular meandering” or just “meandering”. While the eigenvector corresponding to the regular meandering instability was shown to decay radially outward [68], the eigenvector associated with boundary induced drift does not exhibit such decay away from the spiral core. The bifurcations that give rise to the two different instabilities will be termed boundary-Hopf ($H_b$) and meandering-Hopf ($H_m$) respectively. The meandering-Hopf transition causes a quasiperiodic motion of the spiral tip which exhibits complex flower like motions in the domain such as those shown in Fig. 2.8.

Fig. 2.11 shows the bifurcation scenario of the spiral solution in a circular domain with $R = 5$ when $\epsilon$ is used as the control parameter. Other parameters are fixed as before at $a = 0.8$, $b = 0.02$. The stable spiral branch has a Hopf bifurcation (marked as $H_m$ in Fig. 2.11) as $\epsilon$ is increased beyond a critical value where an isolated complex eigenpair cross into the right half plane. A stable branch of meandering spirals arises at this Hopf bifurcation. At a slightly higher value of $\epsilon$, the meandering branch retracts and disappears via another Hopf bifurcation as the isolated pair of eigenvalues returns
2.3 Interaction of Spiral Waves

Fig. 2.11: Bifurcation diagram of a spiral wave solution for $R = 5$, $a = 0.8$, $b = 0.02$. The solid and dashed lines indicate stable and unstable spirals. The filled circles denote quasiperiodic spirals while the unfilled square denotes a spatially uniform state. The symbols $H_b$, $H_m$ and $SN$ stand for boundary-Hopf, meandering-Hopf, and saddle-node bifurcations respectively.

to the left half plane. The spiral solution is then stable in a narrow region in $\epsilon$. The spiral period increases monotonically for increasing $\epsilon$ and the boundaries of the domain start to affect the spiral core. This causes the complex eigenvalue pair associated with the translational symmetry to move away from the imaginary axis into the left half plane. At this point, spirals that are kicked off the center or initiated off-center drift towards the middle of the domain. At the point marked as $H_b$ in Fig. 2.11, the eigenpair associated with translational symmetry crosses the imaginary axis into the unstable right half plane causing the boundary-induced drifting of the spiral. The unstable spiral branch when continued further up in $\epsilon$ turns around in a saddle node bifurcation and terminates at a spatially uniform steady state of the system at some critical value of $\epsilon$.

Next we free both the parameters $\epsilon$ and $R$ in the model and study how the two Hopf transitions are organized in the $(\epsilon, R)$ parameter space. The two parameter bifurcation diagram is displayed in Fig. 2.12, and was assembled by performing many one-parameter continuation runs at constant $R$ and constant $\epsilon$ cuts. The curves corresponding to the meandering-Hopf bifurcation, the boundary-Hopf bifurcation, and saddle-node of spirals are marked as $H_m$, $H_b$, and $SN$ respectively. The regions represent stable rotating spirals, regular meandering spirals, boundary-induced drifting
Fig. 2.12: Two-parameter bifurcation diagram of rotating spiral waves in finite domains. The curves $H_m$, $H_b$, and $SN$ represent the meandering-Hopf bifurcation, the boundary-induced drift related Hopf bifurcation, and saddle-node of spirals respectively. The regions are stable spirals (S), regular meandering spirals (M1), boundary-induced drifting spirals (M2), and no rotating waves (N).
spirals, and no rotating waves respectively. These results show that regular meandering is first stabilized by the influence of the boundary conditions. At still smaller domain sizes, meandering related to the boundary induced drift appears.

2.4 Discussion

In this chapter, we have reviewed research results on complex patterns stemming from the competition and interaction of different structures. In the first section we discussed interfaces between competing traveling and stationary patterns as well as the formation of drifting pattern domains, see [52]. Comparing results in a reaction-diffusion model and in the corresponding amplitude equations showed that drifting pattern domains are stabilized by nonadiabatic effects („interface locking”). The methods used should be helpful in a number of further problems, e.g. in the study of the Turing-wave point in models with differential flow [92].

The second section comprises a treatment of the interaction of the localized structures (solitary pulses) with direct numerical integration and a kinematical approach as well as perturbation theory and numerical bifurcation analysis. Bound pairs have been found to arise from attractive interaction between pulses that manifest itself in a anomalous dispersion, for a a theoretical discussion see [56]. Attractive interaction has also been demonstrated in experiments and realistic model calculation in the catalytic reduction of NO with CO on a Pt(100) surface, see [57]. There, however bound pair formation is prevented by annihilation of one of the partners in contrast to expectation from kinematical theory. The reason for this behavior is the presence of an oscillatory unstable mode of the bound pair solution [62].

A more challenging question is posed by the problem of interaction between spirals resp. topological defects. Here, we have studied the related problem of the interaction of a spiral with a (curved) mirror-wall, exemplified in the strong impact of domain size on the frequency of a rotating wave in small circles. First computational results along these lines have been presented in this thesis. Related experiments and numerical investigations are documented in [83].
3 Spatiotemporal Chaos - Complex Patterns due to Instabilities and Bifurcations

3.1 Spatiotemporal Chaos

Spatiotemporal chaos (STC) is defined by the rapid decay of spatial and temporal correlations. Consequently, the correlation length resp. time are much smaller than the system extension and resp. the observation time. STC manifests itself in the presence of many positive Lyapunov exponents and the proportionality of the attractor dimension to the system volume in 3D (area in 2D, length in 1D) [93]. These criteria have been verified in numerical studies of a number of model systems, e. g. the one-dimensional Kuramoto-Sivashinsky [94] and the complex Ginzburg-Landau-equations [95]. More recently, similar results have been obtained for two-dimensional realistic models of chemical reactions [96] and Rayleigh-Benard convection [97]. STC may emerge for various reasons. First of all, the local reaction kinetics may be chaotic. Diffusion then provides a coupling of different chaotic elements and the question arises whether these elements synchronize or not. Such phenomena have been studied with coupled (chaotic) maps [98] and reaction-diffusion systems with local chaotic dynamics [99, 100].

Here, we constrain ourselves to media with two-variable local dynamics, that may either be oscillatory or excitable (compare Chapter 1). Therein, chaotic patterns often appear in a transition from simple, regular patterns. Here, we study the characteristic properties of these transition in reaction-diffusion systems for spatiotemporal chaos arising from pulse backfiring in 1D and spiral breakup in 2D. In these cases, spatiotem-
Spatiotemporal chaos is well described as a state in which spontaneous creation and mutual annihilation of pulse-like excitation in 1D or topological defects (spiral fragments) in 2D are statistically balanced, but occur in an unpredictable and chaotic fashion. In both cases alternative scenarios have been discovered leading to different phenomenologies of patterns in the transition region. For all examples considered, spatiotemporal chaos resulted from a linear instability of a simple structure like a traveling pulse or a rotating spiral. Thus, other scenarios like the non-normal dynamics responsible for turbulence in simple flow geometries [101] or extremely long-lived chaotic transients towards a final non-chaotic or even periodic attractor [102, 103] could be ruled out for the reaction-diffusion systems studied here. Experimental research has concentrated on spiral breakup scenarios in the Belousov-Zhabotinsky reaction [7, 104, 105, 106]. The two scenarios predicted by the simple activator-inhibitor model studied here are found in these experiment.

3.2 Pulse Bifurcations in Excitable Media

Note: The work in this section contains unpublished material from a collaboration with M. Or-Guil (Dresden) and I. G. Kevrekidis (Princeton). These results supplement the ones presented in [115]. There, we have been concerned mostly with the existence of pulse solutions in the traveling wave ODEs. Here, the emphasis is put on stability of pulse solutions and the way bifurcation can be approximated in the PDE. A longer version has been submitted to Physical Review E.

We investigate the instabilities and bifurcations of traveling pulses in the model excitable medium after Barkley introduced in Chapter 1, see Eqs. (1.4); in particular we discuss three different scenarios for the loss of stability resp. the disappearance of stable pulses. In numerical simulations beyond the instabilities we observe replication of pulses ("backfiring") resulting in complex periodic or spatiotemporally chaotic dynamics as well as modulated traveling pulses. We approximate the linear stability of traveling pulses through computations in a finite albeit large domain with periodic boundary conditions. The critical eigenmodes at the onset of the instabilities are related to the resulting spatiotemporal dynamics and "act" upon the back of the pulses. The first scenario has been analyzed earlier, see [115] for high excitability resp. low excitation threshold: it involves the collision of a stable pulse branch with an unstable pulse branch in a so called T-point. In the frame of traveling wave ordinary differential
equations, pulses correspond to homoclinic orbits and the T-point to a double hetero-
clinic loop. We investigate this transition for a pulse in a domain with finite length
and periodic boundary conditions. Numerical evidence of the proximity of the infinite-
domain T-point in this setup appears in the form of two saddle-node bifurcations.
Alternatively, for intermediate excitation threshold, an entire cascade of saddle-nodes
causing a “spiraling” of the pulse branch appears near the parameter values corre-
responding to the infinite domain T-point. Backfiring appears at the first saddle-node
bifurcation, which limits the existence region of stable pulses. The third case found
in the model for large excitation threshold is an oscillatory instability giving rise to
“breathing”, traveling pulses which periodically vary in width and speed.

3.2.1 Introduction

We briefly review the most important properties of excitable media, which have been
discussed in detail in the previous chapters. One-dimensional excitable media exhibit
nonlinear traveling waves such as wave trains and solitary pulses. Examples include
concentration waves in chemical reactions in solution \cite{107} and on surfaces \cite{42}, signal
propagation in neurons and in cardiac tissue \cite{20}. A pulse is a localized structure; it
may result from a finite amplitude perturbation of a linearly stable rest state. The
pulse shape usually decays exponentially as a function of the distance from the pulse
center. Pulses can be analytically approximated in the asymptotic limit where the
dynamics of the activator is much faster than that of the inhibitor variable, and where
the activator variable diffuses, but the inhibitor does not. There, it can be shown that
a pulse exists and is stable \cite{50}.

More recently, spatiotemporal chaos has been found in a variety of one-dimensional
excitable model systems. Several examples have been reported in models whose kinetics
possess three distinct homogeneous steady states (fixed points) - in our notation type
II excitable media; apart from the stable rest state, these media also exhibit two
unstable fixed points \cite{108, 109, 110, 111}. One example arises in a model for catalytic
CO oxidation, where the inhibitor is a so-called surface structure variable; its kinetic
nullcline displays a sigmoidal shape - threshold dependence on the activator variable
(adsorbate coverage) \cite{24}. In contrast to the standard linear dependence of the inhibitor
nullcline on the activator, this functional form leads to the additional fixed points
mentioned above. Similar behavior is often seen in models in physiology describing the
dynamics of the membrane voltage (activator) controlled by so called gating variables
for the ion channels (inhibitor). The kinetics of these gating variables display again a threshold-type, sigmoidal dependence on the membrane voltage, often leading to three (spatially homogeneous) fixed points for the PDE [20]. If only a single gating variable is involved, these models are qualitatively similar to the equations for catalytic CO oxidation studied here; a good example is provided by the Morris-Lecar model used to describe the membrane potential in a barnacle muscle fiber [23].

If the inhibitor kinetics are fast enough, the CO oxidation model displays an instability that has been colloquially named backfiring of pulses [108]. Related phenomena include the wave-induced chemical chaos found in the Gray-Scott model for an autocatalytic chemical reaction [109], as well as complex behavior in amplitude equations describing dynamics near a Takens-Bogdanov (TB) point [111]. In addition, even reaction-diffusion media whose kinetics are characterized by a single fixed point, sometimes display pulse instabilities and backfiring under excitable conditions [112, 113]. An analysis of the traveling wave ordinary differential equations derived from the original PDEs for the CO oxidation model reveals the basic mechanism for the destruction of stable pulses leading to complex behavior: it involves a so-called T-point, see [115] as well as a rich web of bifurcations of pulse solutions [116]. More recently, a similar bifurcation structure has been found in a model for intracellular calcium waves in pancreatic acinar cells [117] and in the ultrarefractory version of the FitzHugh-Nagumo model [118]. The resulting complex dynamics is often governed by a coherent structure described as a “wave emitting front”, see [115], a nonlinear front involving a spatially uniform unstable state that invades a spatially uniform stable one. The unstable state behind the propagating front evolves into spatiotemporal chaos. Similar behavior has been seen in the amplitude equations in the neighborhood of the TB point, and was named “chaotic nucleation” by those authors [111]. Here, we investigate instabilities and bifurcations in an excitable medium of type II originally derived to describe catalytic CO oxidation [24]. Using a computer-assisted approach we calculate solution branches in large, finite domains, under periodic boundary conditions (which we will, with slight abuse of terminology, refer to as pulses) and perform linear stability analysis of these pulse solutions. The eigenvalues of the corresponding Jacobian reflect the growth exponents of perturbation modes.

A homogeneous steady state of reaction-diffusion equations in an infinite system possesses a continuous spectrum, reflecting the growth rates of perturbation modes. The eigenmodes are harmonic functions, whose wavenumber parametrizes the corresponding eigenvalues. In a very long or infinite system, a single pulse can conceivably
be considered (in the appropriate norm) as a perturbation of this uniform solution. Under these circumstances, the modes far away from the pulse are still delocalized, harmonic wave modes; the continuous spectrum remains essentially unchanged. Furthermore, additional perturbation modes exist that are localized at the site of the pulse and decay exponentially away from it. The corresponding eigenvalues are discrete, in contrast to the continuous band of eigenvalues belonging to the nonlocalized modes. In infinite or periodic systems, the Goldstone mode is an example of such a localized mode. In a spatially homogeneous system it is given by the spatial derivative of the pulse profile, and accounts for a shift in space (due to translational invariance); the corresponding eigenvalue is zero.

A bifurcation or instability of a given solution upon change of a single control parameter is typically accompanied by a single real eigenvalue or a pair of complex conjugate eigenvalues crossing the imaginary axis. For a real eigenvalue, a saddle-node bifurcation is the most common case, while alternative bifurcations like the transcritical or pitchfork bifurcations require certain symmetries for the equations and the solution. The case of a drift pitchfork bifurcation of a stationary pulse was studied in [119]. A destabilizing perturbation grows in an oscillatory manner if the eigenvalues are complex and in a non-oscillatory manner if the corresponding eigenvalue is real. The first case leads to a Hopf bifurcation, while the latter case may correspond either to a saddle-node or to a transcritical or pitchfork bifurcation.

Three examples of dynamics where localized initial conditions beyond the uniform solution excitation threshold do not eventually evolve into stable pulses, are shown in Fig. 3.1. For high excitability resp. small excitation threshold, pulse-like initial conditions evolve in a traveling pulse that splits off new pulse-like excitations traveling in the opposite direction. This is the phenomenon termed backfiring in [108]. It can eventually evolve in spatiotemporally chaotic (I) or complex periodic (II) fashion. For large excitation threshold, modulated traveling pulses are also seen (III). Their shape undergoes a breathing, periodic variation.

### 3.2.2 Model and Methods

We investigate a model of activator-inhibitor type representing a type II excitable medium that was originally derived for CO oxidation on Pt(110) [24]. Type II excitability implies that we work in a parameter regime where the kinetics give rise to one stable and two unstable steady homogeneous solutions. This model is related to
Fig. 3.1: Space-time plots from numerical integration of Eqs. (3.1) showing the time evolution of pulses at parameter values beyond the instability onset. A stable pulse solution for a subcritical parameter value, characterized by rest state $A$, was used as the initial condition. I: Backfiring in the immediate vicinity of a T-point. The resulting behavior is non-periodic for the given system length and initial conditions. II: Backfiring after a saddle-node bifurcation. The resulting behavior is periodic in time for the given system length and initial conditions. III: Modulated traveling waves. The pulse shows a periodic oscillation of shape and speed; variations appear mainly at its trailing edge. Black denotes high values of $u$, white corresponds to $u=0$. Parameters: I: $b=0.07$, $\epsilon=0.1075$, $L=100$, $\Delta T=119.2$, II: $b=0.15$, $\epsilon=0.0931$, $L=100$, $\Delta T=238.42$, III: $b=0.2$, $\epsilon=0.062$, , $L=50$, total integration time $\Delta T=238.42$.

the FitzHugh-Nagumo system and describes the interaction of a fast activator $u$ and a slow inhibitor variable $v$:

$$\begin{align*}
\partial_t u &= \partial_x^2 u + \frac{1}{\epsilon} u(1-u)(u - \frac{b+v}{a}), \\
\partial_t v &= h(u) - v, \\
h(u) &= \begin{cases} 0, & 0 \leq u < 1/3 \\ 1 - 6.75u(u-1)^2, & 1/3 \leq u \leq 1 \\ 1, & 1 < u \end{cases}
\end{align*}$$

(3.1)

with $x \in [0, L]$ and periodic boundary conditions.

The time scales ratio $\epsilon > 0$ is used as the control parameter. The case $\epsilon \to 0$ corresponds to the aforementioned asymptotic limit where stable pulses are expected. The parameter $b$ controls the excitability threshold of the system: the value of $b$ is proportional to the magnitude of the critical perturbation which will trigger a pulse. The value of $a$ is fixed at 0.84 throughout the paper. We will vary $b$, thus varying the excitability. In the parameter range considered here, three relevant fixed points exist: the stable state $A=(0,0)$, the saddle $B=(b/a,0)$, and unstable focus $C$. 
We shall now examine the stability of pulses traveling in a background of the stable rest state $A$. Since we investigate solutions moving with fixed velocity $c$, the analysis of their stability is performed in the comoving frame $z := x - ct$:

$$
\begin{align*}
\partial_t u &= \partial_z^2 u + c \partial_z u + \frac{1}{\epsilon}u(1-u)(u - \frac{b+v}{a}), \\
\partial_t v &= c \partial_z v + h(u) - v.
\end{align*}
$$

(3.2)

In this frame, traveling waves with speed $c$ correspond to time independent, steady solutions. Linearization of these equations around a stationary solution $u_0(z), v_0(z)$ yields an eigenvalue problem for small perturbations $(r(z,t), s(z,t)) \propto (r(z), s(z)) e^{\lambda t}$:

$$
\mathcal{M} \begin{pmatrix} r(z) \\ s(z) \end{pmatrix} = \lambda \begin{pmatrix} r(z) \\ s(z) \end{pmatrix},
$$

$$
\mathcal{M} = \begin{pmatrix} \partial_z^2 + c \partial_z + g_1(z) & g_2(z) \\ \partial_z f(u_0) & c \partial_z - 1 \end{pmatrix}
$$

(3.3)

with

$$
\begin{align*}
g_1(z) &= -\frac{1}{\epsilon} \left( u_0(u_0 - 1) + (u_0 - \frac{b+v_0}{a})(2u_0 - 1) \right), \\
g_2(z) &= \frac{u_0}{\epsilon a}(u_0 - 1).
\end{align*}
$$

The linear stability problem amounts to the determination of the spectrum of the Jacobian $\mathcal{M}$ in (3.3). For the homogeneous steady states $A$ or $B$, at least one of the off-diagonal matrix elements is zero. Thus, the diagonal elements of the matrix $\mathcal{M}$ suffice to compute the spectrum. For the $A$ steady state it is:

$$
\lambda_{A,1} = -\frac{b}{\epsilon a} - k^2 + i\epsilon k, \quad \lambda_{A,2} = -1 + i\epsilon k,
$$

(3.4)

and for the $B$ steady state it is:

$$
\lambda_{B,1} = -\frac{b}{\epsilon a} \left( \frac{b}{a} - 1 \right) - k^2 + i\epsilon k, \quad \lambda_{B,2} = -1 + i\epsilon k,
$$

(3.5)

where $k$ is the wavenumber of the perturbation. In the case of periodic boundary conditions studied here, $k = \frac{n \pi}{L}$ applies. The real part of the eigenvalues $\lambda_2$ is $-1$. The eigenvectors are then $(r,s)^T = (1,0)^T e^{ikz}$ and $(0,1)^T e^{ikz}$.

In general, traveling wave solutions $u_0(z), v_0(z)$ and the eigenfunctions of the Jacobian $\mathcal{M}$ are not available in closed form. Thus, the problem has to be approximated numerically. We approximate pulses by computing steady solutions in a finite length
system and the traveling frame through a pseudospectral discretization of Eqs. (3.2) with periodic boundary conditions and Newton-Raphson iterations. The velocity $c$, which is not known \textit{a priori}, is formally an additional variable along with the Fourier coefficients of the solution. One additional pinning condition singles out one of the infinitely many solutions existing due to translational invariance and allows the numerical computation of the speed. The eigenfunctions and the spectrum of the Jacobian are obtained in Fourier space resulting from a 200 (resp. 250 for case I) mode decomposition of the stationary solution. The zero eigenvalue, which always exists due to the translational symmetry of the problem, is used as a numerical accuracy check and has been obtained with a precision of $10^{-3}$ or better. The time evolution of unstable solutions was computed using an explicit finite difference scheme to solve Eqs. (3.1), discretizing space in 1024 points and using a time step of $\Delta t = 0.0122$.

Because traveling solutions fulfill the conditions $\partial_t u = \partial_t v = 0$ in the comoving frame, they can also be obtained in the traveling wave ordinary differential equations (TWODE) following from Eqs. (3.2):

$$\begin{align*}
\frac{du}{dz} &= w, \\
\frac{dw}{dz} &= -cw + \frac{1}{\epsilon}u(u-1)(u-u_{th}), \\
\frac{dv}{dz} &= (v - h(u))/c,
\end{align*}$$

(3.6)

with $u_{th} = (b + v)/a$. In this frame, a homogeneous solution corresponds to a fixed point, a pulse to a homoclinic orbit, and a front to a heteroclinic orbit. Consequently, in the parameter range studied here, three relevant fixed points exist: $A = (0,0,0)$, $B = (b/a,0,0)$, and the focus $C$.

### 3.2.3 Pulse Bifurcation Scenarios

We consider three cases, at increasing values of the excitation threshold, controlled through the parameter $b$ at fixed $a = 0.84$: Case I ($b = 0.07$), case II ($b = 0.15$) and case III ($b = 0.20$). We proceed as follows: first, branches of pulse solutions on a ring, representative of „true” pulse solutions in an infinite domain, are presented for the relevant range of the control parameter $\epsilon$; we characterize these pulses by their speed $c$ (Fig. 3.2, 3.3b). We then show pulse profiles and spectra at selected values of $\epsilon$ shortly before and after the onset of instability (Figs. 3.3, 3.7, 3.8), and present the destabilizing mode $(r,s)^T$ (Figs. 3.6, 3.7, 3.8). Representative post-instability spatiotemporal
Fig. 3.2: Pulse speed as a function of the parameter $\epsilon$. I: $b = 0.07$, II: $b = 0.15$, and III: $b = 0.2$. Thick lines are pulses with rest state $A$ (i.e. pulses in a large box with periodic boundaries representative of pulses to the state $A$ on the infinite line corresponding to the homoclinic orbits to $A$); thin lines are pulses with rest state $B$. Solid (dashed) lines denote stable (unstable) branches. A $T$ marks our approximation of the double heteroclinic connection point (the T-point) where pulses with rest state $A$ “collide” with pulses with rest state $B$. H denotes Hopf bifurcations, SN denotes saddle-node bifurcations. In the cases II and III, the branch of pulses with rest state $A$ spirals into the T-point. This is not the case for I, compare Fig. 3.3b.
dynamics can be found in Fig. 3.1.

Fig. 3.2 shows pulse speed as a function of the parameter $\epsilon$ for the three cases. The thick lines correspond to stable pulses with background state $A$, while dashed lines correspond to unstable pulses with background state $B$. The transition point between the two families is the so called T-point [124], denoted by a T. This point is defined as a double heteroclinic connection between the fixed points $A$ and $B$ in the frame of the traveling wave ODEs, Eqs. (3.6). In the frame of the original equation (3.1), these heteroclinic orbits correspond to fronts. The branch of pulses corresponding to homoclinic orbits to $A$ in the TWODE may (Fig. 3.2.II, III) or may not (Fig. 3.2.I) spiral into the T-point. Spiraling is observed when two of the eigenvalues of the linearization of the TWODE around the fixed point $B$ computed right at the T-point are complex conjugate.

**Case I**

Fig. 3.2.I shows speed as a function of the parameter $\epsilon$ for $b = 0.07$ computed for pulses in a large system with periodic boundaries. The picture appears at first glance identical with the result found in a continuation of homoclinic orbits in the TWODE presented in [115]. The branch of pulses to $A$ (thick line) does not spiral into the T-point. To be exact, the T-point only exists for pulses on the infinite line; what – at the resolution of our picture – still appears as a T-point will be discussed in more detail below. To understand the nature of the instability in this case, we focus on the solutions near the T-point value $\epsilon_T \approx 0.10744$.

As mentioned above, for an infinite system, the T-point corresponds to a double heteroclinic connection in the traveling wave ODEs, Eqs. (3.6). Close to $\epsilon_T$ the orbits homoclinic to $A$ also approach the fixed point $B$ (and vice versa); this approach to $B$ makes the dynamics in $z$ progressively slower. Thus, close to $\epsilon_T$, pulses with rest state $A$ (resp. $B$) will also exhibit extensive regions close to $B$ (resp. $A$), practically giving rise to fronts between $A$ and $B$ within the pulse profile. When we perform a continuation for a pulse in a ring of finite length, the space on the ring is apart from the excitation plateau divided between residence close to $A$ and residence close to $B$, so that the total period is constant as we vary $\epsilon$.

To study this behavior, numerical continuation techniques are needed. We computed the stationary solutions of Eqs. (3.2) on a ring of length $L = 200$ with 250
Fig. 3.3: a) Four pulse solutions on the ring from the middle branch in (b) illustrating the gradual transformation from solutions identifiable with pulses with rest state $A$ ($u = 0$) to unstable solutions identifiable with pulses with rest state $B$ ($u = 0.083$). The $B$ domain in the waveform becomes wider upon decrease of $\epsilon$, along the middle branch in (b). b) Bifurcation diagram, with respect to $\epsilon$, of pulses for case I, exhibiting two saddle-nodes. The thick solid line can be associated with stable pulses homoclinic to $A$; the thin dashed line can be associated with unstable pulses homoclinic to $B$. The thick dashed line corresponds to the transition region between these two cases; it constitutes the incarnation (for our finite, large ringlength continuation) of the T-point. When the upper saddle-node bifurcation ($SN_1$) takes place, our finite ringlength solution could be still described as an approximation of a pulse to $A$ but with a small $B$ shoulder; the converse description holds at the lower saddle-node bifurcation ($SN_2$). It is reasonable to consider as most representative of the infinite-domain T-point the location, along this thick-dashed line, where the pulse solution contains comparable large patches close to $A$ and close to $B$ – roughly the middle of this middle-branch.
modes, 1024 collocation points and a parameter step size \( \Delta \epsilon = \mathcal{O}(10^{-10}) \). Periodic approximations of pulse solutions with rest state \( A \) are shown in Fig. 3.3a as they approach the T-point and undergo the gradual transition to pulse solutions with rest state \( B \). One can clearly recognize the increasing domain \( B = (b/a, 0) \) at the back of the pulse. As the state \( B \) is unstable, one might expect that the pulse in the ring loses stability as soon as the \( B \) domain gets large enough. However, one should be aware of the fact that the \( B \) domain is moving with the speed of the pulse. It is possible that the \( B \) domain is only convectively unstable in the comoving frame. In other words, the perturbations growing on the \( B \) state in a stationary frame may spread slower than the pulse motion, and the pulse in an infinite domain will be stable. A mathematically precise description of this phenomenon has been given by Sandstede and Scheel [121]. A related result has been obtained by Nii [122], who shows that eigenvalues accumulate in the area bounded by the essential spectra of \( A \) and \( B \). The opposite case, \( i.e. \) perturbations on the \( B \) plateau spread faster than the pulse speed, is described in the next subsection (case II).

Typical spectra are shown in Fig. 3.4 for solutions along the \( c - \epsilon \)-branch. To facilitate a better comparison, the continuous spectra of the rest states \( A \) (Eqs. (3.4)) and \( B \) (Eqs. (3.5)) are depicted as solid lines; the computed eigenvalues are denoted by circles. Fig. 3.4a shows the spectrum of a solution approximating a stable pulse with rest state \( A \), while Fig. 3.4f shows that of a solution approximating an unstable pulse with rest state \( B \). In both cases, the parameter \( \epsilon \) is “far” from the T-point bifurcation and the eigenvalues belonging to nonlocalized eigenvectors compare well with the continuous spectrum of the respective rest state. Figs. 3.4b-e show the gradual transition between the two cases. Fig. 3.4b shows the spectrum for a stable solution for which the domain \( B \) is just starting to appear in the back of the pulse. Its width is \( L_B \approx 5 \). Several discrete eigenvalues have appeared on the negative real axis; a similar spectrum has been predicted and found by Sandstede and Scheel [121].

Gradually, as the \( B \) plateau grows, two distinct phases of spectrum movement are observed. First this new \( B \)-associated parabola moves to the right in the complex plane and at some point it starts crossing the imaginary axis. That is precisely the first saddle-node bifurcation we observe - the critical eigenvalue is the tip of this secondary parabola, whose origin we just discussed, see Fig. 3.4c. When both \( A \)- and \( B \)-plateaus are equally present, one expects to see echoes of both \( A \)- and \( B \)-continuous spectra in the solution spectrum, and that is indeed seen in Fig.3.4d. Near the lower saddle-node bifurcation, the \( B \)-plateau is almost as wide as the system length. There, one may
Fig. 3.4: Spectra of pulses in case I for different values of $\epsilon$. The solid lines correspond to the continuous spectrum of the rest state $A$, Eqs. (3.4) (left parabola) resp. rest state $B$ (right parabola), Eqs. (3.5). The pictures show the typical change of the spectrum along the solution branch $c$-$\epsilon$. Parameters: $a = 0.84$, $b = 0.07$, $L = 200$ ($L = 400$ for picture d), a) $\epsilon = 0.106425824$, b) $\epsilon = 0.107446965$, c) $\epsilon = 0.107446995547$, d) $\epsilon = 0.1079$, e) $\epsilon = 0.107446995527$, f) $\epsilon = 0.109936154$.

describe the solution more reasonably as an approximation to a pulse with rest state $B$ with a short $A$ domain at its front. The eigenvalue spectrum at this situation is depicted in Fig. 3.4e. The second (lower) turning point occurs when the “discretized” parabola of eigenvalues that corresponded initially to continuous spectrum of $A$ moves to the right and the real eigenvalue at the tip of this discretized parabola crosses the imaginary axis. Fig. 3.4f shows the spectrum for a solution approximating a pulse with rest state $B$ far from the T-point bifurcation for a high values of $\epsilon$.

Focusing on the first instability ($SN_1$), the critical eigenmode corresponding to the saddle-node appears localized at the back of the pulse (see Fig. 3.5). In addition, it is acting mainly on the activator (Fig. 3.5b); we observe that new pulses can now split off the existing pulse (“backfiring”). Fig. 3.6 shows the time evolution beyond the saddle-node bifurcation in a comoving frame. The initial condition is the stable pulse just before the bifurcation. Fig. 3.6a shows how a perturbation with support over the $B$-plateau grows with time, while the shape of the preceding front essentially does not change. The back (what starts as the $B$-plateau) grows until its maximum reaches the excited state $u = 1$. The excited domain widens, but its plateau is unstable against
oscillating perturbations. Therefore, the situation rapidly evolves, and the growing excited domain breaks down, giving rise to two new seeds for $A$-pulse like entities. This behavior has been described by the term “backfiring” [108]. Backfiring occurs repeatedly, and the newly generated pulse-like structures annihilate upon collision with similar objects traveling in the opposite direction; this interplay of instability, new pulse generation and annihilation gives rise to the non-periodic space-time behavior shown in Fig. 3.1.I.

Altogether, our results give an interesting and well resolved picture of stability of pulses on a large finite ring near the T-point. In contrast, earlier computations as in [115] and in [116] have used coarser steps in the continuation algorithms and smaller domains. The present computations show how the pulse solutions change in a gradual fashion; and that two distinct solutions branches show an extremely narrow hysteresis; the middle branch mediates the transition between the two different pulse types. The main changes appear in an extremely tiny region of parameter space and can therefore only be captured with careful numerics.
Fig. 3.6: Time evolution for a value of $\epsilon$ after the saddle-node bifurcation $SN_1$ in a comoving frame. The initial condition is the stable pulse-like solution just before the bifurcation (thick black line in a)). The thin lines in a) demonstrate how the shape of the solution changes only at the back of the pulse-like solution. A localized perturbation grows in amplitude and width in the course of time and approaches the rest state $C$, where $u = 1$. Further evolution of the dynamics is given by the dashed line in picture b). Due to oscillatory instability of the homogeneous state $C$, the corresponding domain cannot become large. A breakdown leads to the generation of two further pulse-like states traveling in opposite directions (thin solid line).
Fig. 3.7: Case II: a) Eigenvalues at $\epsilon = 0.0927$ before a saddle-node bifurcation of a pulse decaying into the rest state $A$; b) eigenvalues at $\epsilon = 0.09299$ after this bifurcation. A discrete eigenvalue crosses the imaginary axis. c) Unstable solution and d) corresponding destabilizing eigenmode $(r, S)^T$ at $\epsilon = 0.09299$. Parameter: $a = 0.84$, $b = 0.15$, $L = 100$.

Case II

Fig. 3.2.II shows the $c - \epsilon$ diagram for $b = 0.15$. As in case I, we again observe here the transformation from pulses with rest state $A$ to pulses with rest state $B$ at what appears like a T-point. The branch corresponding to “pulses to $A$” spirals into this T-point. This behavior is caused by imaginary eigenvalues of the fixed point $B$ in the TWODE at the T-point conditions, and has been predicted from general arguments [124]. Thus, the branch of initially stable pulses with rest state $A$ undergoes a sequence of saddle-node bifurcations upon approaching the T-point. This sequence of saddle-node bifurcations is consistent with the results of Sandstede and Scheel [121]. The spectrum of $A$-pulses near the T-point in the infinite system approaches the union of the essential spectrum of $A$ and the absolute spectrum of $B$. In this case, however, the absolute spectrum of $B$ contains real positive eigenvalues. One may, therefore expect infinitely many saddle-node bifurcations as the T-point is approached. In the stability-relevant first saddle-node bifurcation, the solution branch of stable wavetrains with rest state $A$ turns around and becomes unstable. As is required for a saddle-node bifurcation, a single real eigenvalue crosses the imaginary axis. This can be seen in Fig. 3.7, left column. The two spectra correspond to pulse-like solutions before and after the saddle-node bifurcation along the solution branch. The unstable pulse and the destabilizing eigenmode are shown in Fig. 3.7, right column. Once more, the destabilizing mode affects primarily the back of the pulse. It is worth noting, that upon further continuation the branch of pulses to $A$ spirals towards the T-point related situation over a cascade of saddle-node bifurcations. Each saddle-node bifurcations adds an additional positive eigenvalue to the spectrum.
3.2 Pulse Bifurcations in Excitable Media

Numerical simulations of the model Eqs. (3.1) shortly after the saddle-node bifurcation, for values of $\epsilon$ for which no solution with rest state $A$ exists, exhibits the phenomenon we termed above “backfiring”. In the transients, our pulse-like object generates near its back other pulse-like entities traveling in the opposite direction, see Fig. 3.1.II. After this transient period, the resulting spatiotemporal pattern in our finite domain becomes periodic in time. Simulations show, though, that this observation depends on the initial conditions: non-periodic patterns like the one shown in Fig. 3.1.I may also appear for the same parameter values.

Case III

Fig. 3.2.III shows the $c - \epsilon$ diagram for $b = 0.2$. Here, the pulse solution with rest state $A$ becomes unstable through a Hopf bifurcation even before the first saddle-node bifurcation is reached. The eigenvalue spectrum is shown in Fig. 3.8, left column, on both sides of this Hopf bifurcation. It can be seen that one discrete pair of complex conjugate eigenvalues crosses the imaginary axis. The second column of Fig. 3.8 shows the pulse-like solution after the bifurcation as well as the real and imaginary parts of the critical eigenmode $(r, s)^T$. Note that the perturbation due to the eigenmode is localized and has its main contribution – once more – at the back of the pulse. As the corresponding eigenvalues are complex conjugate, the resulting perturbation oscillates with time at a frequency given by the imaginary part of the eigenvalues.

Numerical integration of the model equations (3.1) shows that the Hopf bifurcation which leads to destabilization is supercritical [116]. The resulting pattern after desta-
bilization consists of a *modulated* traveling pulse which oscillates in time, especially at its back (compare the simulation shown in Fig. 3.1.III.)
3.2.4 Conclusions

We have investigated the transition from stable pulse propagation to various regimes of more complicated spatiotemporal dynamics, namely modulated pulses, periodic and chaotic pulse backfiring. In all three cases, the transition can be explained by either a Hopf instability (modulated pulses) or a saddle-node bifurcation (leading to backfiring) of the stable pulse solution. In all cases, the transition is connected with either a single or a pair of complex conjugate discrete eigenvalue(s) with zero real part(s). In a finite domain with periodic boundary conditions – the typical experimental setup for investigation of pulses – spectra change continuously near the T-point in the fashion described in case I. The form of the corresponding critical eigenmode(s) allows some insight into how pulses become unstable resp. evolve in space and time. The dynamics in general still contains mostly propagating localized pulse-like structures whose evolution is governed by the unstable eigenmode(s) in the Hopf case (modulated) or by the critical eigenmode of the saddle-node bifurcation. Typically, the critical eigenmodes have support at the back of the pulse.

The results here should carry over to models with similar phenomenology mentioned in the introduction. Preliminary results [125] show that the transition to wave-induced chemical chaos in the Gray-Scott model [109] is also caused by a saddle-node bifurcation of pulses near a T-point. T-points can only be found in systems with multiple homogeneous fixed points (e.g. one stable rest state and two additional unstable steady states). The complex behavior seen in media with a single stable fixed point [112, 113] may be caused through a different mechanism. For a model of the catalytic NO-CO reaction, upon change of control parameter first modulated traveling waves are seen and then periodic backfiring is found. This cannot be due to a T-point, but may instead be caused by a global bifurcation of the periodic modulated pulses - a scenario already suggested in a study of the present model with different control parameters [116]. Finally, it is important to note that a simple instability of a finite wavelength pulse solution, like the Hopf bifurcation in case III, only leads to a modulation of the shape, while a saddle-node bifurcation limits the existence of a certain type of pulses and may give rise to replication of pulse-like structures (cases I, II). The role of saddle-node bifurcations in the replication of pulses and creation of space-time defects has recently been investigated in the Gray-Scott model [126] and in the Complex-Ginzburg-Landau equation studied in the next section and in [127].
3.3 The Transition from Phase to Defect Chaos

**Note:** The following passage is a modified version of the introduction of a long paper, submitted to Physica D, corresponding to the Letter in A.6 and is joint work with the same team of authors.

The transition from *phase to defect chaos* for the one dimensional complex Ginzburg-Landau equation (CGLE) was recently related to the bifurcation properties of a family of coherent structures called *modulated amplitude waves* (MAWs), see [127]. In many cases these patterns show an erratic behavior in space and time: such behavior is commonly referred to as *spatiotemporal chaos* [1, 93, 130]. Examples of extended systems displaying such chaotic dynamics in one spatial dimension include: heated wire convection [131], printers instability and film drag experiments [132, 133], eutectic growth [134], binary convection [135], sidewall convection [136], the far field of spiral waves in the Belousov-Zhabotinsky reaction [7, 104, 105, 106], the Taylor-Dean system [137], hydrothermal [138, 139] and internal [140] waves excited by the Marangoni effect and the oscillatory instability of a Rayleigh-Bénard convection pattern [141]. Near the pattern forming threshold, the dynamics of such systems can often be described by so-called amplitude equations.
Fig. 3.9: Values of the phase gradient peak as a function of the interpeak spacing. The curves denote a typical bifurcation diagram of MAWs and the shaded area schematically indicates the typical values for near-MAW structures during the large scale chaotic dynamics of the CGLE. Coefficients are $c_1 = 0.65$, $c_3 = 2$ and correspond to transient phase chaos as observed in Fig. 3.10 (f,i,j). Hence the shaded area reaches $P_{SN}$ just before defects appear. Arrows show the typical evolutions of near-MAWs.
Fig. 3.10: Summary of our main results and picture for the formation of defects from phase chaotic states. If interpeak spacings \( p \) become larger than the biggest period for MAWs \( P_{SN} \) then defects are formed. (a,b) Example of a coherent structure, phase gradient and modulus of a MAW at \( c_1 = 0.6, c_3 = 2, P = 30 \). (c) Space time plot showing the stable propagation of the MAW from (a,b) in a small system of size \( P \) with periodic boundary conditions. (d) The same MAW as initial condition creates defects at \( c_1 = 0.7, c_3 = 2 \) where \( P > P_{SN} = 26.8 \). All space time plots show the phase gradient encoded in grayscale (minima appear dark, maxima bright). Black bars above the x-axis denote the size of \( P_{SN} \) specific to the parameters of the panel. (e,g,h) Large scale chaos at \( c_1 = 0.63, c_3 = 2, L = 512 \). (e) Snapshot of the phase gradient profile with individual interpeak spacings \( p \). (g) Space time evolution of phase chaos and (h) distribution \( D(p) \) showing \( p \ll P_{SN} \) and no defects. A transient of \( t \approx 10^4 \) is not shown. (f,i,j) Large scale chaos at \( c_1 = 0.65, c_3 = 2, L = 512 \). (f) Snapshot of the phase gradient profile \( t = 120 \) before the first defect forms and the MAW (dotted, \( P = P_{SN} \) overlayed onto the long structure. (i) Transient phase chaos with a fast and long structure travelling through the system and defect chaos nucleating at \( t = 400, x = 360 \). This structure was shown in (f) and in (i) again a transient of \( t \approx 10^4 \) is not shown. (j) The tail of the distribution of \( p \) reaches \( p > P_{SN} \) which denotes the long structure that leads to the break down of phase chaos. The distribution \( D(p) \) shown in (h) is also reported (dashed line). From the comparison of the two it is evident that the distributions do not modify dramatically, while \( P_{SN} \) decreases noticeably.
When the pattern forming bifurcation from the homogeneous state is a forward Hopf bifurcation, the appropriate amplitude equation is the CGLE [1], which in one spatial dimension reads as:

$$\frac{\partial_t}{\partial_x} A = A + (1 + i c_1) \frac{\partial^2}{\partial_x^2} A - (1 - i c_3) |A|^2 A , \quad (3.7)$$

where $c_1$ and $c_3$ are real coefficients and the field $A = A(x, t)$ has complex values.

For different choices of the coefficients numerical investigations of the CGLE have revealed the existence of various steady and spatiotemporally chaotic states [1, 127, 128, 129, 130, 141, 142, 143, 144, 145, 146, 147, 148, 149, 150, 151, 152, 153, 155]. Many of these states appear to consist of individual structures with well defined propagation and interaction properties. It is thus plausible to use these structures as building blocks for a better understanding of spatiotemporal chaos.

As a function of the coefficients $c_1$ and $c_3$, the CGLE (3.7) can exhibit two qualitatively different spatiotemporal chaotic states known as phase chaos (when the modulus $|A|$ is at any time bounded away from zero) and defect chaos (when $|A|$ can vanish leading to phase singularities). It is under dispute whether the transition from phase to defect chaos is sharp or not, and if a pure phase-chaotic (i.e. defect-free) state can persist in the thermodynamic limit [146, 154]. We will address these issues by suggesting a mechanism for the formation of defects related to the range of existence of MAWs.

The emerging picture for defect creation is outlined in the following and illustrated in Figs. 3.9-3.11. (i) Our investigation starts with the study of MAWs, which are uniformly propagating, spatially periodic solutions of the CGLE. These MAWs are parametrized by the average phase gradient $\nu$ and their spatial period $P$. Our study is confined to the case $\nu = 0$ for reasons specified below. A bifurcation diagram of MAWs with varying spatial period $P$ at fixed coefficients $c_1, c_3$ is shown in Fig. 3.9. Spatial profiles and the stable propagation of a particular MAW are presented in Fig. 3.10a-c. Isolated MAW structures consisting of just one spatial period $P$ play an important role in defect formation. In particular, for fixed CGLE coefficients the range of existence of coherent MAWs is limited by a saddle-node ($SN$) bifurcation which occurs when $P$ reaches a maximal period $P_{SN}$ (see Fig. 3.9).

In Fig. 3.10 black bars at the $x$-axis denote the respective size of $P_{SN}$. (ii) If the MAWs are driven into conditions with $P > P_{SN}$ a dynamical instability occurs leading to the formation of defects (Fig. 3.10d). (iii) Slowly evolving structures reminiscent of MAWs (“near-MAWs”) are observed in the phase chaotic regime (Fig. 3.9...
and Fig. 3.10e,f). In order to characterize such states, we have examined the distribution \( D(p) \) of spacings \( p \) between neighboring peaks of the phase-gradient profile. In particular for sufficiently long spacing \( p \), the observed phase chaos structures are often very similar to a single period of a coherent MAW (Fig. 3.10f). (iv) When a phase chaotic state displays spacings \( p \) larger than \( P_{SN} \), phase chaos breaks down and defects are formed (at \( t = 400, x = 360 \) in Fig. 3.10i). Thus, the MAW with \( P = P_{SN} \) may be viewed as a critical nucleus for the creation of defects. Defect creation in phase chaos is similar to the dynamical process by which isolated MAW structures generate defects (Fig. 3.10 (d)). Therefore purely phase chaotic states are those for which \( p \) remains bounded below \( P_{SN} \) (Fig. 3.10 (g)), while defect chaos can occur when \( p \) becomes larger than \( P_{SN} \) (Fig. 3.10 (i)). (v) A more detailed study of the probability distribution of the \( p \)'s shows that for large \( p \) the probability decays exponentially (Fig. 3.10 (h,j)).

As long as \( P_{SN} \) has a finite value, we expect that, possibly after a very long transient time, phase chaos will eventually be replaced by defect chaos. However, in a finite domain of the phase chaotic region, MAWs of arbitrarily large \( P \) exist: we expect that in this region, even in the thermodynamic limit, phase chaos will persist. Fig. 3.11 shows the main quadrant of the CGLE coefficient space. The region of persistent phase chaos is bounded by the Benjamin-Feir-Newell curve (thin dot-dashed) and the curve along which \( P_{SN} \to 1 \) (full curve).

More details can be found in [127]. There, we study systematically bifurcation diagrams of the MAWs (see Fig. 3.9), starting from the homogeneous oscillation. The MAW range of existence is limited by a saddle-node bifurcation in which two branches of MAWs merge and disappear. Moreover, we have studied the incoherent dynamics of near-MAW structures, and have shown that for \( p > P_{SN} \), i.e. beyond the saddle-node bifurcation, near-MAWs evolve to defects. Furthermore, we have studied various aspects of spatio-temporal chaos in the CGLE, and relate the observed continuous \((L_1)\) and discontinuous \((L_3)\) transitions (see Fig. 3.11) to properties of the MAWs. In the phase chaotic regime, near-MAWs with various periods are created and annihilated perpetually. The transition to defect chaos takes place when near-MAWs with periods larger than \( P_{SN} \) occur in a phase chaotic state. We have argued that the saddle-node curve for \( P_{SN} \to 1 \) is a lower bound (see Fig. 3.11) for the transition from phase chaos to defect chaos, see also [127]. The typical dynamics of the phase chaotic regime can be related to the competition of two instabilities of the MAWs, splitting and attractive interactions. It is, moreover, possible to give a reasonably good estimate for the numerically measured transition from phase to defect chaos by considering these
linear stability properties of MAWs. The numerical values for the transition are shown as circles in Fig. 3.11, whereas the estimate from the stability analysis of MAWs is given by the thin dotted line. The estimate is obtained as follows: MAWs may become unstable to splitting modes that decrease $p$ and hence prevent defect nucleation at small values of $c_1c_3$ or to attractive interaction modes that increase $p$ and therefore favor defect nucleation at large values of $c_1c_3$. The thin dotted line shows the transition from dominance of splitting to dominance of interaction modes. The instabilities are illustrated in [127].

Here, the analysis was restricted to MAWs with $\nu = 0$. A related study has been done for $\nu \neq 0$ MAWs. This objects are relevant for the understanding of superspirals that have been found recently [7]; an example is shown in Fig. 3.12a. Far away from the core, superspiral approach MAWs. They stem from the modulational Eckhaus instability. Far into the Eckhaus unstable regime, superspirals breakup, see Fig. 3.12b. This phenomenon is the 2D analogue of the defect creation discussed in this section and is described in great detail in the next section. However, we will elaborate below that superspiral breakup in 2D, similar to defect creation in 1D, is related to the saddle-node bifurcation of MAWs with non-zero winding number $\nu \neq 0$, where $\nu$ corresponds to the selected (unstable) wavenumber of the spiral.
Fig. 3.11: Phase diagram of the CGLE, showing the Benjamin-Feir-Newell curve (thin dot-dashed) where the transition from $\nu = 0$ waves to phase chaos takes place. The curves $L_1$ (long dashed), $L_2$ (thin dashed) and $L_3$ (dashed) as obtained in [142, 144] separate the various chaotic states. The circles correspond to our data for the $L_1$ and $L_3$ transition along the 17 cuts in coefficient space that we studied. The filled circles show our data for the observed transition to defect chaos, while the open circles correspond to the location where $p_{max} = P_{SN}$. Only small discrepancies between these two can be observed. Finally the full curve shows the $P_{SN} \to \infty$ limit which we believe to be a lower boundary for the transition from phase to defect chaos. The thin dotted line gives the parameter values for which the real parts of the eigenvalues for the splitting and interaction instabilities are equal.
Fig. 3.12: Experimental example for (a) modulated amplitude waves and (b) spiral breakup due to an Eckhaus instability in the Belousov-Zhabotinsk reaction, after [104]
3.4 Spiral Breakup in Excitable Media

Note: Subsections 3.4.1 - 3.4.4 comprise a shorter updated version of the review on spiral breakup published originally with M. Or-Guil and M. Falcke (MPI Dresden) and listed as paper i) in Appendix A.16. Subsections 3.4.5 - 3.4.7 are newly written for this thesis.

3.4.1 Introduction

Rotating spiral waves are frequently observed in homogeneously and heterogeneously catalyzed chemical reactions [12, 42] and various biological systems, namely slime mold aggregation [156], cardiac tissue and calcium waves in frog eggs [20]. Pattern forming reaction diffusion systems can be classified as either oscillatory, excitable or bistable media with either none, one or two linearly stable homogeneous states [50, 55]. Patterns form either due to the instability of a steady state in oscillatory media or due to suprathreshold, finite amplitude perturbations of homogeneous stable steady states in bistable and excitable media. In the latter case, they have a stable rest state and respond to a suprathreshold perturbation with an amplification followed by saturation and recovery. The existence of an excitation threshold is a generic feature. A popular model for oscillatory systems is the complex Ginzburg-Landau equation (CGLE) that is suited to describe media near a Hopf bifurcation [1]. So called activator-inhibitor systems are widely used to model excitable media [157].

In this section, we analyze an instability of rotating spirals that leads to spatiotemporally chaotic dynamics [93] – the so called spiral breakup. The problem has received considerable attention for various reasons. For instance, the observation of “defect-mediated” turbulence in numerical simulations of the CGLE has been attributed to the breakup of spirals [29, 158, 159, 160, 161, 162]. Until the early nineties, it was unclear if spatiotemporal chaos or irregular activity is possible in homogeneous two-dimensional (2D) continuous excitable media. The first observation of chaotic wave patterns were reported in discrete models [163, 164]. The availability of faster computers led to the discovery that various models exhibit spatiotemporal chaos. Among them are models of cardiac tissue [165, 166, 167, 168] and activator-inhibitor models of FitzHugh-Nagumo type designed to capture essential aspects of pattern formation [169, 170, 171, 172].

Most experimental systems that exhibit spirals are considered to be excitable or
oscillatory. An important motivation for the study of excitable media has been the quest for the cause of irregular electrical activity in cardiac muscle [173]. Experiments in thin sheets of heart tissue displayed only stable spirals in contrast to the irregular activity seen in experiments with whole hearts [174]. Consequently, it has been suggested that irregular activity in the heart might be a genuinely three-dimensional phenomenon [175]. Thus more realistic three-dimensional, anisotropic models of the heart and excitable media have been investigated and revealed various sources of irregular activity on the surface including intricate dynamics of scroll waves [176, 177] and the analogue of breakup in three dimensions [178, 179]. The reason for the onset of ventricular fibrillation as well as possible treatments still remains a subject of intense experimental and theoretical research [180]. In pattern forming chemical reactions, progress in the design of open reactors has finally also yielded experimental results that demonstrate a controlled transition to spatiotemporal chaos via spiral breakup in the Belousov-Zhabotinsky reaction [104]. Additional examples of transitions from spiral patterns to irregular spatial organization have been also reported in catalytic surface reactions [183].

In reaction-diffusion media, two different mechanism of spiral breakup exist. Spirals break because the waves emitted from the spirals center (core) are either destabilized by transverse perturbations that appear only for fast inhibitor diffusion [184, 185, 186, 187] or by unstable modes in the radial direction [165, 166, 167, 168, 169, 170, 171, 172, 188, 189, 190]. The examples mentioned above belong solely to the latter case. In what follows we shall concentrate only on destabilization against modes in the radial direction.

It is important to note that in all the mentioned examples of spiral breakup in models and experiments two different scenarios are observed – spirals may break first close to their center or alternatively far away from the core [190]. Breakup near the center is found in most simulations in excitable media [167, 169, 171], while breakup far away from the core is typically seen under oscillatory conditions both in chemical experiments [104] and in simulations of the CGLE [160, 161]. One exception from this rule of thumb is provided by recent studies of a model of intracellular calcium waves where breakup far away from the center is observed under excitable conditions [191].

Here, we will present results on a simple activator-inhibitor model that exhibits both types of breakup depending on the chosen control parameters [171, 190]. After we have discussed the phenomenology of breakup in these models we will turn to a stability analysis of periodic waves in the two model systems. The rationale of the treatment
of periodic waves is as follows: stable rotating spirals do emit periodic waves, and far away from the core the concentration patterns resemble planar periodic wavetrains. If these wavetrains are unstable, breakup in the radial direction may result.

For the CGLE it is possible to compute the stability of periodic wave solution analytically, because the waves and the corresponding eigenfunction have the form of Fourier-modes [159]. For given parameters, periodic waves in the CGLE become unstable below a certain wavelength through an Eckhaus instability. If the most stable wave number tends to zero, one uses the term Benjamin-Feir instability. It indicates an instability of the spatially homogeneous oscillations. Eckhaus instabilities first appear through long wavelength perturbations that lead to spatially slowly varying modulations of the periodic waves. In the nonlinear stage, Eckhaus unstable wavetrains may evolve to the modulated amplitude waves (MAWs) discussed in the previous section. In the CGLE, the Eckhaus instability is of convective nature and several groups have pointed out that spiral breakup requires an absolute instability of the emitted wave train [159, 160, 161]. Consequently, stability results should be further analyzed with respect to the distinction of absolute and convective instability.

In excitable media, stability analysis has been originally restricted to kinematic treatment of the phenomenon of alternans [188, 189]. Alternans is a period doubling instability of periodic waves, wherein the width of excitation pulses oscillates between two different values. It has been identified to play an important role in the process of breakup in models of cardiac tissue like the Beeler-Reuter model and some simplifications [165, 167]. When an analytic stability analysis is not feasible, the reduction to an one-dimensional (1D) problem may be applied. The stability of one-dimensional wavetrains is then analyzed numerically with sufficient accuracy. This approach has the advantage that it is applicable to most reaction-diffusion models. Thus it is also helpful in the case of realistic models for which studies have been mostly constrained to numerical simulations of the dynamics so far. We will demonstrate its usefulness in the analysis of our two example systems below.

Altogether, the reduction of the spiral stability problem near breakup to the stability analysis of one-dimensional wavetrains gives important insight but fails to completely resolve the puzzle. The reason for potential discrepancies is given by nonlinear behavior not captured by the linear stability analysis or ingredients of the 2D dynamics like spiral meandering [55, 192] that do not have a counterpart in one dimension. This leaves two alternatives – on the one hand it is possible to extend stability calculations to two dimensions, on the other hand one may study an analogue of the breakup
instability in one dimension.

Analytical approaches to spiral stability in 2D are limited to the case of very sharp interfaces resp. large separation of time-scales of the activator and inhibitor [193]. Numerical stability analysis has been crucial in the understanding of the meander instability of rotating spirals. It was identified as a Hopf bifurcation that introduces a second frequency apart from the rotation frequency in the spiral movement [68]. The spiral tip no longer follows a circular trajectory during spiral meandering, but instead gives rise to flowerlike trajectories. However, numerical stability analysis in two dimensions is computationally expensive and in practice still restricted to rather small domain sizes (typically up to system sizes of a few spiral wavelength) as well as to the iterative computation of the largest eigenvalues. It has also been shown that the domain size can change the properties and the stability of the spiral in experiments and simulations [83]. In these computations, the spiral core starts to interact with the boundaries if the system is too small. Depending on the parameters, the critical domain radius where the boundary notably starts to affect the spiral dynamics is between half and one spiral wavelength. Stability computations have also been performed in one of the models discussed but so far have not been able to resolve the cause of the spiral instability at breakup, see the section on spiral interaction in the previous chapter. The reasons for this failure lie both in the rather strong interaction of the spiral with the boundaries for breakup parameters and in the nature of the breakup instability that will become clear below.

The second possibility, reduction to an analog 1D problem, has been advocated by Tobias and coworkers [161]. They showed that, for the CGLE, patterns similar to the breakup simulation in 2D can be observed in 1D sources of periodic waves. The periodic waves emitted from the 1D source become unstable if they select a wavenumber, that would be absolutely unstable in an unbounded system. Their studies and the results given below also imply that the core structure of the spiral is not important, but that the instability mainly depends on the selected wavelength of the source in 1D or the spiral in 2D.

Thus, we will study 1D sources and demonstrate that the different scenarios of breakup can be reproduced in the 1D analog. Finally, we compare results from numerical stability analysis of planar wavetrains with simulations of breakup in 1D and 2D and give an overview over recent results.
3.4.2 The Modified Barkley (MB) Model

To study spiral breakup, we use the 2D version of the model described above in the section about pulse instabilities. We recall its most important features: It describes the interaction of a fast activator \(u\) and a slow inhibitor \(v\) variable:

\[
\begin{align*}
\frac{\partial u}{\partial t} &= -\frac{1}{\epsilon} u(u-1)\left(u - \frac{b + v}{a}\right) + \Delta u, \\
\frac{\partial v}{\partial t} &= h(u) - v, \quad (3.8)
\end{align*}
\]

The form of \(h(u)\) describes a delayed production of the inhibitor and the equations have been used to model patterns in a catalytic surface reaction [24]. The change of \(h(u)\) from the standard choice \(h(u) = u\) [25] lead to the possibility of spatiotemporal chaos in 2D due to spiral breakup for \(\epsilon > \epsilon_{BU}\) [96, 171].

The parameter choice \(a < 1\) yields excitable (oscillatory) behavior for \(b > 0\) \((b < 0)\) and \(0 < \epsilon \ll 1\). In both cases, an unstable focus exists with \((u, v) = (u_0, v_0)\) in the local dynamics of Eqs. . In the excitable case, two more fixed points appear: \((u, v) = (0, 0)\) is the stable rest state and \((u, v) = (b/a, 0)\) is a saddle that marks the threshold of the excitable medium. Throughout this section, \(a\) is fixed to 0.84 and \(b\) and \(\epsilon\) are varied.

For simulations in 2D, zero-flux boundary conditions have been employed.

Simulations have been performed in 2D as well as in 1D reductions that allow for a better comparison with stability results obtained in 1D. All stability computations have been performed in 1D systems with periodic boundary conditions. The methods are the same as in the section about pulse instabilities. Spiral dynamics is found from numerical integration and is investigated for a large number of values of the control parameters. Accordingly, wavetrains and related patterns are studied in the 1D form of the equations. For small \(\epsilon\), the system settles into stable rotating resp. traveling waves.

If the wave is stable, all eigenvalues \(\omega_j\) from the numerical stability analysis possess nonpositive real parts. Bifurcations take place, if one or more eigenvalues cross the imaginary axis. For two component systems, discretization with either \(N\) nodes or \(N\) Fourier modes leads to \(2N\) algebraic equations with \(2N + 1\) unknowns. The equations are then supplemented by a so called pinning condition, that fixes the position of the
wave and thus selects one of the equivalent solutions from the family of waves that is given due to the translational invariance of the system.

### 3.4.3 Phenomenologies of Spiral Breakup

In this section, results of numerical simulations for the two models are described and compared to recent experiments in a chemical reaction. We have investigated the dynamics of spiral waves as a function of the excitation threshold parameter $b$ and the ratio $\epsilon$ of the timescales of activator $u$ and inhibitor $v$. The model is excitable for small positive values of $b$ and oscillatory for small negative values of $b$ as long as $\epsilon \ll 1$. Almost independent of the choice of $b$, we find a transition from rotating spiral waves to spiral breakup at $\epsilon$-values between 0.07 and 0.08 [171] in both the excitable and oscillatory regimes [171, 190]. The simulations are usually started with a spiral initial condition that has been obtained at nearby values of $\epsilon$. The first run has been performed at $\epsilon = 0.02$ and then $\epsilon$ is increased in small steps ($\Delta \epsilon = 0.0025$) until destabilization occurs. In the vicinity of the spiral breakup instability we have used much finer “resolution” in $\epsilon$. The destabilization of spirals just above the critical $\epsilon$ however depends on the choice of $b$. For the given model we find two different scenarios that are exemplified in Fig. 3.13. For excitable conditions, spiral breakup appears first close to the center and the irregular pattern spreads then outward (cf. Fig. 3.13). For oscillatory conditions, we observe a different behavior. Spirals first break up far away from the center and eventually relatively large spiral fragments surrounded by a “turbulent” bath remain (cf. Fig. 3.13b). The size of the surviving part of the spiral shrinks if $\epsilon$ is further increased until finally no trace of the initial spiral persists in the long run (Fig. 3.15a). At closer inspection we observe that, even inside the surviving part of the initial spiral, modulations in wavelength and period appear. The amplitude of these modulation grows with growing distance to the core. Fig. 3.15b gives the minimum and maximum periods measured as a function of the distance to the spiral center for parameter values corresponding to the patterns in the first four frames of Fig. 3.15a. It is obvious that with increasing $\epsilon$ the modulation grows faster and thus the critical modulation amplitude necessary to cause breakup is reached at distances closer to the center. In addition, the breakup in the excitable regime is usually preceded by a meander instability of the rotating spirals, while for oscillatory conditions breakup bounds a region of stable spiral rotation [171]. The breakup far
away from the core has been also observed in experiments in the Belousov-Zhabotinsky reaction, [104] and in numerical simulations of the CGLE [160, 161, 104]. More recent studies provide also an experimental verification of the core breakup scenario, see Fig. 3.14 taken from [7].

As discussed in the introduction, further insight on the nature of the spiral instability may be obtained by a reduction to one spatial dimension. Here, we demonstrate this procedure for the MB model. Therein, a 1D wave source is studied as an analogue of a spiral in 2D. We will show that the essential properties of the breakup instability depend mostly on the asymptotic selected wavetrain and not on the detailed structure of the source (e.g. the spiral core geometry or the center of a target pattern). Results for 1D sources and 2D spirals should allow for a quantitative comparison if their selected wavelengths are the same. A 1D source of periodic waves is given by the equations

\[
\begin{align*}
\frac{\partial u}{\partial t} & = -\frac{1}{\epsilon}u(u-1)(u - \frac{b+v}{a}) + \frac{\partial^2 u}{\partial r^2} + \frac{G(r)}{r} \frac{\partial u}{\partial r}, \\
\frac{\partial v}{\partial t} & = h(u) - v.
\end{align*}
\] (3.9)

Dirichlet \((u(0) = u_1)\) and zero-flux \((\partial u(L)/\partial r = 0)\) boundary conditions are used at respective ends of the 1D system of length \(L\). With a choice \(G(r) = 1\), Eqs. (3.9) represents the radial part of Eqs. (3.8) and the last term on the right hand side describes the impact of curvature. The Dirichlet boundary at \(r = 0\) is a source of waves similar to the core of the spiral in 2D. Note however, that the 1D source is not equivalent to the spiral in 2D. It rather describes the radial dynamics of a target pattern emitting circular waves. By changing \(u_1\) the wavelength of the emitted wavetrain can be varied. Here, we picked \(u_1\) values that select the same wavelength as spirals in 2D just below the breakup instability. For simplicity, we neglect curvature effects and set \(G(r) = 0\).

Space-time plots from the integration of Eqs. (3.9) are presented in Fig. 3.16 both for oscillatory and excitable conditions. In both cases, the wave train emitted from the left boundary at \(r = 0\) exhibit an instability upon increase of \(\epsilon\) at a critical value \(\epsilon_C\). In the excitable case with \(b = 0.07\), the instability appears always very close to the source (see Fig. 3.16a). For \(b = -0.045\) we find a scenario similar to the behavior in 2D (cf. Fig. 3.16b). Breakup occurs first far away from the Dirichlet boundary, i.e. the source of waves. Upon increase of \(\epsilon\) the breakup of the wavetrain moves towards the source until it gets to a distance of about one wavelength. The wavelength \(\lambda_{dir}\) and period \(\tau_{dir}\) of the waves selected by the Dirichlet boundary is for \(\epsilon\)-values near the breakup almost
the same as the selected wavelength $\lambda_0$ and period $\tau_0$ of the spirals in 2D in line with earlier results in the CGLE [159]. Nevertheless, the critical value for the 1D source $\epsilon_C \approx 0.081 > \epsilon_{BU} \approx 0.074$. The transition between the two scenarios happens around $b = 0.04$. The values of $\epsilon_C$ are not very sensitive to changes in $b$ and are slightly above 0.08. Thus, we find the counterpart of breakup near and far away from the spiral core in the 1D model. In particular, the appearance of different qualitative scenarios does not depend on specific 2D ingredients like meandering and curvature.
Fig. 3.13: Two different scenarios of spiral breakup shown at different stages in time. Both scenarios lead to irregular dynamics. (a) The breakup appears first close to the center and spreads then outward. This breakup mechanism is related to an absolute instability of the one-dimensional periodic wavetrain. 
(b) The breakup appears first far away from the center. At the end, a stable spiral fragment with finite radius is left, surrounded by a “turbulent” bath. This breakup mechanism is related to a convective instability of the one-dimensional periodic wavetrain. The figures show simulations of the model, Eqs. (3.8). Greyscale: Black resp. white corresponds to \( u \)-values of 1 resp. 0. Parameters: (a) \( \epsilon = 0.08, b = 0.07, a = 0.84 \); (b) \( \epsilon = 0.0752, b = -0.045, a = 0.84 \); system size: 100 \( \times \) 100; zero-flux boundary conditions
Fig. 3.14: Experimental example for core breakup in the Belousov-Zhabotinsky reaction, after [7]
Fig. 3.15: A closer inspection of the spiral breakup mechanism shown in Fig. 3.13. (a) Frames 1-6 show snapshots of the final dynamics for increasing values of $\epsilon$. The spiral is stable for subcritical values of $\epsilon$ (first frame). For supercritical values of $\epsilon$, only a fragment of the spiral remains stable. The radius of this fragment decreases with increasing $\epsilon$ (frames 2-5). Finally, the spiral fragment disappears (frame 6). (b) Minimum and maximum periods of the waves of a spiral fragment in dependence on the distance to the fragment core $r - r_0$ for different values of $\epsilon$. The values of $\epsilon$ correspond to those of the frames 2-5 in (a). The higher the values of $\epsilon$, the stronger the modulation. Parameters: $b = 0.07, a = 0.84$, and in (a) $\epsilon = 0.073$ (frame 1), $\epsilon = 0.074$ (2), $\epsilon = 0.0746$ (3), $\epsilon = 0.0752$ (4), $\epsilon = 0.076$ (5), $\epsilon = 0.077$ (6). The system size is again $100 \times 100$. 
Fig. 3.16: One-dimensional space-time plots of the MB model, Eqs. (3.9), showing instabilities of the emitted wave train which are related to the spiral breakup mechanism. Space is depicted horizontally, time from top to bottom. Black represents high values of the variable \( u \). Dirichlet boundary conditions are used in the left hand side as a source of waves (cf. text), while zero-flux conditions apply to the right side. (a) The breakup of wave trains occurs near the source at a critical value of \( \epsilon \). This instability mechanism resembles thus the spiral breakup in Fig. 3.13a. (b) The breakup of waves occurs first far away from the wave source. Upon increase of \( \epsilon \), the breakup site moves towards the left, Dirichlet boundary. This instability mechanism resembles thus the spiral breakup in Fig. 3.13b. Parameters: system length \( L = 100 \), \( G(r) = 0 \), \( a = 0.84 \) and (a) \( \epsilon = 0.082 \), \( b = 0.07 \); (b) \( \epsilon = 0.0815 \) (frame 1), \( \epsilon = 0.082 \) (2), \( \epsilon = 0.083 \) (3), and \( b = -0.045 \).
3.4.4 Stability Analysis of Spirals near Breakup: Scaling Arguments and Numerical Bifurcation Results

The basic argument in spiral stability analysis is quite simple. The spiral selects for a given set of parameters a particular wavelength $\lambda_0$ and related temporal period $\tau_0$. At the same moment, there exists a one-parameter family of periodic wave solutions with varying speed $c$, temporal period $\tau$ and wavelength $\lambda = c\tau$. At small wavelength, the wavetrains either exhibit an instability (Eckhaus in the CGLE, alternans in cardiac tissue models) or cease to exist (saddle node or drift pitchfork bifurcations in FitzHugh-Nagumo systems). These minimum stable wavelengths and periods shall be called $\lambda_{min}$ respectively $\tau_{min}$ in the following. In a few cases (CGLE [159], Rinzel-Keller model [194]) it is possible to compute the instability or bifurcation at $\tau_{min}$ analytically. In other cases, kinematic theories (alternans, [188, 189]) or approximations by singular perturbation theory (excitable media) may yield useful information on the nature of this instability. For the models treated here, none of these methods are sufficient to yield an understanding of the instability at $\tau_{min}$. This calls for alternative methods, namely numerical stability analysis. Here, we will present qualitative results on spiral stability obtained from scaling arguments for the MB model and then present quantitative results found by numerical stability computations.

A. Scaling Arguments

For excitable media given by reaction-diffusion equations of the form presented in Eqs. (3.8), a large body of theoretical work has been done in the limit of very small $\epsilon$. In particular, scaling laws originally predicted by Fife [195] have been explicitly derived for equations of the type used here by Karma [196] with the constraint $\epsilon^{1/3} \ll 1$. This result can be directly applied to our model and yields the following relation:

$$\tau_0 = 2\pi \left( \frac{aB}{\sqrt{2} \left( a/2 - b \right) \left( 1 - a/2 + b \right)} \right)^{2/3} \epsilon^{1/3}, \quad (3.10)$$

where $B = 1.738$ and $a, b$ and $\epsilon$ are the control parameters. By use of singular perturbation theory [157] one can also derive an approximation for the dispersion relation $c(\tau)$. In lowest order, one usually obtains that $\tau$ goes to $0$ as $c$ goes to $0$. A correction that includes the contribution of the front and back interfaces to the period of the wavetrain yields a minimum period where the dispersion curve turns around in a
saddle-node bifurcation [24]. This minimum period for the MB model is given by
\[ \tau_{\text{min}} = 16 \left( \frac{a}{(a - 2b)(2 - a + 2b)} \right)^{1/2} \epsilon^{1/2}. \] (3.11)

Results of numerical simulations resp. numerical stability analysis at small \( \epsilon \) show indeed an algebraic dependence of the quantities \( \tau_0 \) and \( \tau_{\text{min}} \) of the form
\[ \tau_{\text{min}} = b_0 \epsilon^\beta, \quad \tau_0 = a_0 \epsilon^\alpha. \] (3.12)

Such a scaling prevails up to \( \epsilon \)-values of 0.06 for the example studied here. The scaling exponents and prefactors differ significantly from the analytic predictions. For the prefactors we find \( b_0 = 19.74 \) (\( b_0 = 20.05 \)) and for the exponents \( \beta = 0.613 \) (\( \beta = 0.612 \)) for a value of \( b = 0.07 \) (\( b = -0.045 \)).

The analytic predictions from Eq. (3.11) are \( \beta = 1/2 \) and \( b_0 = 15.37 \) (\( b_0 = 13.51 \)) for a value of \( b = 0.07 \) (\( b = -0.045 \)). Similarly, we obtain from simulations of spiral dynamics in 2D, prefactors of \( a_0 = 13.66 \) (\( a_0 = 13.70 \)) and exponents \( \alpha = 0.398 \) and \( \alpha = 0.417 \) for a value of \( b = 0.07 \) (\( b = -0.045 \)). The analytical formula Eq. (3.10) predicts \( \alpha = 1/3 \) and \( a_0 = 17.22 \) (\( a_0 = 16.23 \)) for a value of \( b = 0.07 \) (\( b = -0.045 \)). Overall, both analytical and numerical data display a faster growth of \( \tau_0 \) compared to \( \tau_{\text{min}} \) with increasing \( \epsilon \). Thus, one should expect an instability at large enough \( \epsilon \) where \( \tau_0 \) and \( \tau_{\text{min}} \) become equal. If we exploit the two analytic expressions or extrapolate the numerical scaling found at \( \epsilon < 0.06 \), breakup should occur at values of \( \epsilon \approx 1 \). As demonstrated in the previous section, breakup happens much earlier for \( \epsilon \) values between 0.07 and 0.08. This is related in particular to the complete breakdown of scaling for the spiral period for \( \epsilon > 0.06 \). Altogether, it is not too surprising that the scaling arguments fail quantitatively, because the breakup occurs at values of \( \epsilon \) much bigger than the ones assumed in the derivation of the Eqs. (3.10) and (3.11). Scaling arguments have been used with more success in the spiral breakup in cardiac tissue, where the instability appears upon decrease of \( \epsilon \) and the scaling properties of the spiral waves are not in line with the Fife scaling.

**B. Numerical stability results**

In the following, we will compute the stability of wavetrains along the dispersion curve directly by numerical stability analysis using the approach described above. Consequently, we apply periodic boundary conditions and consider \( N \) pulses on a ring. The
evolution of perturbations of a spatially periodic, traveling wave $\mathbf{U}_0(z)$ is described by $\mathbf{W}_{jn}(z)e^{\omega_j nt}$, where $\mathbf{W}_{jn}$ and $\omega_{jn}$ are eigenvectors and eigenvalues obtained from linear stability analysis. The indices $j, n$ will be specified below. There are infinitely many eigenvalues and eigenfunctions. For $L = \infty$, $\omega_{jn}$ are located on continuous curves in the complex plane. The solution is stable when all eigenvalues $\omega_{jn}$ have negative real parts. The ring length $L \text{ resp.}$ the wavelength $\lambda = L/N$ are used as bifurcation parameters. In addition, the ring length $L$ and the spatial discretization of the equations ($\Delta z$ for finite differences or $k_{max}$ for spectral methods) determine the quality of the approximation to the continuous spectrum of the periodic waves. If the method provides the necessary accuracy, it yields information on the minimum stable period $\tau_{min}$ for the given medium as well as insight in the nature of the corresponding instability, the eigenvalue spectra and the eigenfunctions describing the dynamics of perturbations to the wavetrains. These results are then compared to the periods of spirals $\tau_0$ and 1D sources $\tau_{dir}$ near the breakup threshold and the simple idea about selection of an unstable rotation period is tested and discussed.

If we consider periodic traveling waves with constant shape $\bar{\mathbf{U}}_0(z)$ and constant speed $c$ in 1D, a few facts on the eigenvalues and the eigenfunctions are known a priori. Since our media are homogeneous and translation invariant, any translation of a given periodic solution is also a valid solutions with identical stability properties. This translational symmetry of the wavetrains is reflected by an eigenvector $\mathbf{W}_{00} = d\mathbf{U}_0/dz$ with zero eigenvalue $\omega_{00} = 0$ (Goldstone mode). Symmetry arguments require the eigenfunctions of the periodic operator, obtained by linearization around a wavetrain with wavelength $\lambda$, to be Bloch functions $\mathbf{W}_{jn}(z) = e^{i2\pi n z/L}\Phi_{jn}(z)$ with $\Phi_{jn}(z) = \Phi_{jn}(z + \lambda)$ and $n = 0, \ldots, N - 1$ [48]. The above Bloch form can be used in an Ansatz that reduces the stability problem for the infinite domain to a stability problem in a domain of length $L$ with the wavenumber $k$ as an additional parameter. For the problems considered here, we simply considered sufficiently many pulses on the ring. The number of pulses $N$ and the corresponding ring length $L$ in our approach do correspond to the step length of the modulation wavenumber $k$ in the mentioned bifurcation analysis based on the Bloch form. Thus, the finite length $L$ of the ring in our analysis imposes a “quantization” of $2\pi/L$ on the wavenumbers of the eigenfunctions. For a wavetrain with given wavelength $\lambda$, this finite size effect can be reduced by increasing the number $N$ of pulses where $L = N\lambda$. To achieve reasonable convergence of the stability properties of the wavetrains, basic solutions with $N > 8$ pulses on the ring have to be used for the two models under consideration. Usage of only a single pulse results in
underestimation of $\tau_{\min}$ by 10-15 percent.

The eigenvectors corresponding to the eigenvalues with the largest real part are modulations of the Goldstone mode of the approximate form $W_{0n} \approx e^{i(2\pi n/L)z}W_{00}(z)$. The amplitudes of the eigenfunctions are largest in the fronts and backs of the pulses in the wavetrain in contrast to the Fourier eigenfunctions in the CGLE [159]. Thus, the fastest growing modes correspond to an alternating compression and expansion of subsequent pulses in line with the observations in the 2D simulations of Fig. 3.13 and experiments in the Belousov-Zhabotinsky reaction [104]. The calculations for both models reveal an instability reminiscent of the Eckhaus instability in the CGLE. As the length of the ring is shortened, the spectra of the wavetrains shift towards larger real parts and cross the imaginary axis at $L_{\text{min}} = N\lambda_{\text{min}}(N)$. At $\tau = \tau_{\text{min}}$ resp. $\lambda = \lambda_{\text{min}}$, the leading part of the spectrum switches from a parabola opening to the left and touching zero to a symmetric curve with two positive maxima of the real part of $\omega_{jn}$ at nonzero imaginary values.

The dispersion curves describe the dependency between speed $c$, wavelength $\lambda$ and period $\tau$. It is sufficient to plot two of three quantities, since $\lambda = c\tau$. Here, we plot $c$ vs. $\lambda$ and display stable (unstable) wavetrain solutions along the dispersion curve with full (dashed) lines. The dispersion curve (see Fig. 3.17) ends towards small wavelength and periods at a nonzero value of the speed $c$ and not at a saddle-node bifurcation or at zero velocity as known from standard excitable media [55, 194]. Closer inspection reveals that the wavetrains instead bifurcate off one of the additional unstable equilibria with zero amplitude. Such a scenario may be typical for excitable media with additional unstable fixed points and is of course prevented for models which just have the rest state of the medium as a fixed point. The Eckhaus-type instability, however, appears somewhere along the dispersion curve and cannot be deducted from the form of the dispersion curve alone (as could be a saddle-node bifurcation). The instability happens just before the speed has a local minimum along the dispersion curve. The goal of the previous analysis has been to provide a comparison between the minimum stable period $\tau_{\text{min}}$ and the selected periods $\tau_0$ and $\tau_{\text{dir}}$ found in simulations near the spiral breakup instability and its one-dimensional analogue.

We have made the comparison mainly between the results of the simulations with the Dirichlet source and the stability analysis of wavetrains. Fig. 3.18 shows the comparison between $\tau_{\text{min}}$ (thick dashed line, squares) and $\tau_{\text{dir}}$, the selected period of the Dirichlet source (thick full line, triangles). The results are plotted in double logarithmic format clearly indicating the algebraic dependence of $\tau_0$ and $\tau_{\text{min}}$ on the
Fig. 3.17: Dispersion curves describing the dependency between the velocity $c$ and the wavelength $\lambda$ of a periodic one-dimensional wavetrain for different values of $\epsilon$ for the MB model. The velocity was normalized by the velocity $c_0$ of a single pulse in an infinite system. The dashed lines mark the unstable range. Parameters: $a = 0.84, b = 0.07$

parameter $\epsilon$ as long as $\epsilon < 0.06$. In addition, we have displayed earlier results on the minimum period computed in simulations with a single pulse on a ring [171] (thin dashed lines, diamonds). One clearly sees the deviation due to the discretization of the possible wavenumbers of the eigenfunctions that results in an underestimation of $\tau_{\text{min}}$ by about ten percent. Furthermore we have displayed the spiral periods for the respective parameters (thin full lines, circles). The upper panel of Fig. 3.18 shows the data for the oscillatory case ($b = -0.045$). Again, there is a narrow interval where stable 1D sources and spirals exist with $\tau_0$ resp. $\tau_{\text{dir}} < \tau_{\text{min}}$. This is presumably related to the convective nature of the Eckhaus instability. Consider that convective instabilities in the infinite system will be suppressed for fixed (Dirichlet) boundary conditions [197]. Perturbations are then simply advected out of the system boundaries. The radial dynamics of spirals in 2D are analog to the Dirichlet case; the amplitude of the waves in the spiral goes to zero in the center of rotation. This corresponds to a “self-imposed” Dirichlet boundary condition. We also note that the spiral breakup appears at lower values of $\epsilon$ than the instability of the Dirichlet source despite the fact that in both cases practically the same period and wavelength are selected. This is most likely due to a nonlinear effects not captured by the linear stability analysis.
3.4 Spiral Breakup in Excitable Media

Fig. 3.18: Dependence of the period \( \tau \) on the parameter \( \epsilon \) for the MB model, Eqs. (3.9). \( \tau_{\text{min}} \) denotes the minimum period for which a periodic wave train is stable in an infinite system. \( \tau_{\text{dir}} \) denotes the period selected by a Dirichlet boundary condition acting as a wave source. \( \tau_0 \) is the period selected by the corresponding spiral. The diamonds denote the minimum period computed in simulations with a single pulse on a ring. Parameter: \( b = -0.045 \) (upper panel), \( b = 0.07 \) (lower panel), and \( a = 0.84 \).

For the excitable case (breakup close to the center, \( b = 0.07 \)) shown in the lower panel of Fig. 3.18, the simple stability argument eventually works. At the instability of the 1D source, the period produced by the source equals the minimum stable period of wavetrains: \( \tau_{\text{dir}} = \tau_{\text{min}} \). While this is in line with naive expectations, the result is somewhat surprising at second glance. It basically suggests that the instability does not strongly depend on the boundary conditions, which is typical for an absolute instability. This is not to be expected for Eckhaus instabilities of traveling waves. The stability analysis of a traveling wave yields an eigenvalue problem with an operator without reflectional symmetry due to the \( \partial U / \partial z \)-term and the asymmetric shape of the traveling wave. Therefore, one expects that perturbations to the right (in direction of propagation) and to the left (against the direction of propagation of the periodic waves) should behave differently. Thus, it is very unlikely that the fastest growing perturbations have zero group velocity as is the case for problems with reflection symmetry \( z \leftrightarrow -z \).
3.4.5 Recent Developments

In the previous sections we have established the existence of two different scenario of spiral breakup and showed that core breakup practically coincides with the instability of planar wavetrains, while far field breakup probably requires an absolute instability of the planar wavetrain similar to the scenario originally proposed for the CGLE [159]. Recent efforts have yielded a more substantial mathematical analysis of convective and absolute instabilities and their relation to boundary conditions [123]. Application of these results to the example described above shows that the CGLE arguments hold indeed also for the MB model [198]. In the same work, the situation for core breakup turned out to be less clear. One difficulty is that new defects once created in the far field tend to be quite robust and are usually not pushed towards the boundary, even if the instability is linearly convective. This leads typically to deviations between the predictions from linear stability and two-dimensional simulations already noticed in extensive studies of the CGLE [144]. As we showed above, a reduction to one-dimensional sources allows for more accurate predictions.

However, in recent simulations with another modified Barkley model (the nonlinearity in the inhibitor dynamics of Eqs. (3.8) had been replaced by the simpler function $h(u) = u^3$). The impact of the nonlinear dynamics is suppressed for a core breakup scenario in two dimensions [199]. The comparison to the numerical stability analysis of wave trains confirmed the picture described above and in [190]. A subsequent analysis of the propagation direction of perturbations and the nature of the wavetrain instability revealed the following picture [199]. For the case of an Eckhaus instability, perturbations are always traveling away from the core (convective instability) in line with the reasoning by Sandstede and Scheel [198]. The window between the Eckhaus and corresponding absolute instability may however be very tiny and below the resolution of the numerical results presented above and in [190]. Similar to the results in [198], the group velocity of the perturbation is also much smaller than in case of the „convective”, far-field breakup. Altogether such a convective Eckhaus instability gives phenomenologically rise to core breakup. Quantitative differences in the group velocity at the convective Eckhaus instability lead to a qualitative change in the breakup scenario. A more recent study of the $u^3$-model show an interesting alternative: the Eckhaus instability is replaced by an instability to short wavelength modes. These modes can be shown to propagate inward and therefore cause a core breakup [199]. These recent results suggest an extension of the argument by Kramer and Aranson
that requires an absolute instability of the emitted wavetrains. **Spiral breakup occurs if destabilizing modes propagate towards the core.** For the convective Eckhaus instability, the critical modes travel away from the core. Consequently, the first unstable modes that travel towards the core appear, if the instability becomes absolute and the original argument does not contradict our new criterion. The outcome is different in the case of a convective instability with modes that propagate inward, then no absolute instability is required and core breakup can be expected right at the onset of the (short wavelength) instability [199].

A further complication in the analysis of spiral breakup is spiral meandering. Meandering leads to a Doppler effect in the waves emitted from the spiral core. Due to the modulation of the spiral tip curvature a motion of the source of waves results in a modulation of the wavelength and frequency of the emitted wave trains. Originally, this effect has been analyzed in simulations with core breakup [171]. Meandering spirals can, however, also display far field breakup as has been demonstrated also with simulations of the Atri model for intracellular calcium waves [191]. The influence of meandering and the resulting Doppler effect in core breakup has recently been verified in experiments with the Belousov-Zhabotinsky reaction [105]. Far field breakup of (presumably) meandering spirals with a non-decaying modulation of the wavetrain in the far field („superspirals“ [201]) have been studied in the same reaction [7, 106]. Here, an interpretation of the breakup in terms of the nonlinear consequences of the Eckhaus instability is possible already within the CGLE [200]: similar to the case of the Benjamin-Feir instability analyzed in the previous section, modulated amplitude waves arise from the Eckhaus instability of a wave train with a given wavenumber $q$. Their average winding number is $\nu = q$. The spatial period of the modulation is typically limited by a maximum value $P_{SN}$. Meandering excites the convective instability of the outgoing wave trains that is usually suppressed due to the fixed boundary conditions. If the resulting modulated wavetrains have $P > P_{SN}$ far field breakup results. If, on the contrary, $P < P_{SN}$ a modulated amplitude wave in the far-field is found.

### 3.4.6 Characterization of Spiral Defect Chaos

So far we have been concerned with the transition from regular spiral waves to irregular spiral defect chaos. Another relevant issue is the characterization of spatio-temporal chaos. First of all, the chaotic nature of the dynamics can be established by the computations of Lyapunov exponents and the subsequent estimation of the Kaplan-Yorke
dimension of the attractor [202]. The emerging pattern can be called spatiotemporally chaotic if (a) positive Lyapunov exponents appear and (b) if the attractor dimension scales extensively with the length (1D), the area (2D) resp. the volume (3D) of the system studied [1, 93]. This has been achieved for various systems: coupled map lattices [94], the 1D CGLE [95, 146], the 1D MB model exhibiting backfiring [203], the 2D CGLE [204] and the Navier-Stokes equation [97]. For the present system in 2D, the explicit computation of Lyapunov exponents and the proof for extensive scaling of the attractor dimension has been achieved by Strain and Greenside [96].

The interpretation of the attractor dimension in spatiotemporal chaos lacks the simple geometric intuition of the fractal dimension of attractors in low dimensional chaotic systems. From the results presented here, it is plausible to compare attractor dimensions with the number of unstable coherent structures that may be viewed as „building blocks“ for spatiotemporal chaos. For the two-dimensional spiral defect chaos under consideration, the number of topological defects is a relevant quantity. Its changes in time as well as the spatial correlation between topological defects has been investigated beyond the breakup instability, see [205]. The main result is that defects just above the breakup threshold are still relatively stable and exhibit only small fluctuations of the defect number around its mean. This can be explained by the strong anticorrelation between defects - defects avoid being close to each other (defect liquid). Far above threshold, the variance of the defect number becomes equal to the mean and the anticorrelation vanishes. This behavior is expected for randomly distributed topological defect (defect gas). Interestingly, the transition from defect liquid to defect gas is accompanied by a notable increase of the attraction dimension density [96].

3.4.7 Conclusions

We have investigated reaction-diffusion models exhibiting spiral breakup and its one-dimensional analogue with various phenomenologically different scenarios. Altogether, we can distinguish four different routes to breakup - far field and core breakup with and without meandering, respectively. Simple far field breakup is seen in the CGLE, in the MB model as well as in the first experiments with the BZ reaction [104]. Simple core breakup is so far found only in the $u^3$ model. Both scenarios are found for a wave emitting source triggered by suitable boundary conditions in 1D in the MB model, compare [190]. The Doppler induced core breakup caused by spiral meandering is
seen in simulations [171] and recent experiments with the BZ reaction [105]. Far field breakup with meandering, finally, has been found in the Atri model for intracellular calcium waves [191] and offers a good explanations for the latest observations of spiral breakup in the BZ reaction [106, 7].

Scaling arguments account for qualitative predictions of the breakup instability in the MB model. The parameter of interest in the breakup instability is the ratio of time scales of inhibitor and activator \( \epsilon \). The cases of breakup mentioned here are in general related to instabilities of the wave trains emitted from the spiral core in the radial directions. Our studies generalize the earlier conjecture by Aranson and Kramer [159]: spiral breakup requires that the emitted wave train is unstable and that some of the unstable perturbations are transported towards the core. Hence, breakup does not require an absolute instability of the wavetrain; a convective instability that transports towards the spiral core is sufficient as well. Stability analysis of wavetrains in 1D reveals that the long wavelength, modulational Eckhaus instability appears in many models below a minimum stable wavelength \( \lambda_{\text{min}} \). Only recently, a scenario with a short wavelength instability [199] has been discovered in the \( u^3 \) model. The phenomenological distinction between core and far field breakup is not reflected in a similar clear distinction on the level of linear stability analysis. Far field breakup usually is connected with the Eckhaus instability, where the critical modes are always transported away from the core [198] and the group velocity of these modes is relatively large. Then, breakup requires the absolute instability, i. e. some modes have to be transported towards the core. However, an Eckhaus instability with a group velocity near zero may appear as core breakup as the case of the MB model shows [190, 198]. Alternatively, shortwavelength modes that are transported towards the core may trigger spiral breakup [199].

Here, numerical stability analysis has been restricted to the analysis of traveling waves in one dimension. Future work may extend the numerical method to two and more dimensions or to modulated traveling waves and time periodic patterns. Such a project poses a considerable challenge to numerical methodology, but promises a resolution of some unsettled issues. The possibility of core modes or the existence of modulated amplitude wavetrains in realistic reaction-diffusion models could be investigated by such an approach. Another missing piece in the story of spiral breakup is a quantitative modeling of the extensive experimental material in the Belousov-Zhabotinsky reaction by Flesselles et al. [104] as well as the clarification of related experimental observations in other chemical and biological reactions.
3.5 Discussion

This chapter dealt with the onset of spatiotemporal chaos in excitable and oscillatory media as well as with transitions between different phases of spatiotemporal chaos. In addition, a method for the characterization of spatiotemporal chaos is suggested and tested in an excitable medium. The common thread is (a) that spatiotemporal chaos results from instabilities of simple patterns or coherent structures like pulses, spirals or wavetrains and (b) that spatiotemporal chaos is determined by the unstable coherent structures emerging from instabilities and bifurcation of these simpler patterns.

In one dimension, stable pulses in excitable media with a rest state and two additional unstable, homogeneous steady states exhibit a so called T-Point bifurcation, where the stable pulse branches collides with an unstable pulse branch. This transition is truly discontinuous in the infinite medium, see [115]. If the pulse on the infinite line is approximated by a pulse on a ring, the transition becomes continuous and is in fact replaced by a narrow hysteresis bounded by two saddle-node bifurcations. For different parameters, pulses exhibit saddle-node bifurcation or Hopf bifurcation near the T-point, which limit their existence region or render them unstable. In all cases, eventually spatiotemporal chaos by backfiring appears as can be already predicted by the critical eigenvectors of the stability analysis.

In two dimensions, spiral breakup is now established as a common phenomenon in excitable and oscillatory media. The present work shows that there are four different possible scenarios, core breakup with and without meandering and far field breakup with and without meandering. A stability analysis of simple core and far field breakup is provided for a reaction-diffusion model in [190]. It is shown that breakup is related to an Eckhaus instability of the outgoing wavetrain.

In one dimensional oscillatory media two different chaotic states namely phase and amplitude chaos are observed. Amplitude chaos is characterized by the presence of space-time defects. We have identified the critical nucleus for defect creation by investigating modulated amplitude waves (MAWs) resulting from Eckhaus- and Benjamin-Feir instabilities in [127]. MAW analysis also yields a bound for defect nucleation from phase chaos establishing that phase chaos may persist in the limit of large system size and observation times.

Finally, we characterized two-dimensional space time chaos resulting from spiral breakup by a statistics of spiral fragments (topological defects). These defects display
liquid-like behavior near respective gas-like behavior far from the breakup instability, see [205]. The respective pair correlation functions determine the ratio of variance to mean value of defects, this ratio is much smaller than 1 for the liquid and 1 for the gas.
4 Complex Patterns in Anisotropic and Heterogeneous Media

4.1 Introduction

So far we have considered complex pattern formation in systems with a high degree of internal symmetry. These media have been assumed to be homogeneous in space and stationary in time. Thus, the underlying equations have translation and rotation symmetry (in two and three dimensions) in space as well as translation symmetry in time. In addition, the effect of the boundaries has been mostly neglected by using large systems and/or periodic boundary conditions. Here, we show that removal of one of these symmetries leads to new phenomena. In the first section, anisotropic media are studied where the rotational symmetry is broken due to intrinsic properties of the media (like crystal structure in catalysis or tissue structure in biological systems). Anisotropy may cause square-shaped patterns as well as growing fragments traveling along a preferred direction. The suppression of labyrinthine patterns and the emergence of so called stratified space-time chaos is caused by anisotropy. The second section deals with the impact of system geometry (sharp corners or inert inclusions) as well as with the impact of heterogeneity on structure formation. Emphasis is put on localized and periodic heterogeneities. The interaction between intrinsic length scales of the pattern and the externally imposed length scale of the heterogeneities in the periodic case turns out to be crucial for pattern dynamics in heterogeneous media.
4.2 Diffusion Anisotropy

4.2.1 Introductory Remarks

Recently, there has been considerable interest in the emergence of dissipative structures in systems with broken rotational symmetry, namely convection in liquid crystals [206] and chemical waves in catalytic surface reactions [42]. Experimental and theoretical studies of such anisotropic systems showed novel phenomena like ordered arrays of topological defects [207], anisotropic phase turbulence [208], reaction-diffusion waves with sharp corners, see [209, 210] and [211], as well as traveling wave fragments along a preferred orientation [212]. Anisotropy is also often present in pattern formation processes in biological media, e.g. in cardiac tissue [20, 215]. In reaction-diffusion systems, anisotropy usually enters via the diffusion constants. In two dimensions, we can distinguish „simple” and „complex” anisotropy. Simple anisotropy can usually be removed by simple scale transformations to dimensionless units:

\[
\frac{D_x}{\partial x^2} + \frac{D_y}{\partial y^2} \rightarrow \frac{\partial^2 u}{\partial x'^2} + \frac{\partial^2 u}{\partial y'^2} = \Delta' u,
\]

where \( x' = x/\sqrt{D_x} \) and \( y' = y/\sqrt{D_y} \). Apart from boundary effects, a medium with simple anisotropy cannot be distinguished from the isotropic system. New phenomena require a complex anisotropy that cannot be removed by simple scaling. A realization is a medium with two interacting chemical species with concentrations \( u \) and \( v \) that both diffuse with different anisotropy rates. By scaling, we can still make one of the diffusion operators isotropic, e.g. by setting \( x' = x/\sqrt{D_{v,x}} \) and \( y' = y/\sqrt{D_{v,y}} \).

\[
D_{u,x} \frac{\partial^2 u}{\partial x^2} + D_{u,y} \frac{\partial^2 u}{\partial y^2} \rightarrow \delta^{-1} \Delta' u + \delta^{-1} d \frac{\partial^2 u}{\partial y'^2}
\]

\[
D_{v,x} \frac{\partial^2 v}{\partial x^2} + D_{v,y} \frac{\partial^2 v}{\partial y^2} \rightarrow \Delta' v
\]

The effective anisotropy is now contained in the parameter \( d = \delta D_{u,y}/D_{v,y} - 1 \), where \( \delta = D_{v,x}/D_{u,x} \).

Another variant are nonlinear diffusion terms, i.e. the diffusion coefficient depend explicitly on the concentrations. If these functional dependencies in different directions are different, scaling may restore isotropy for specific values of the concentrations, but leaves the medium effectively anisotropic. This case is relevant for surface reactions and
4.2 Diusion Anisotropy

Fig. 4.1: Examples for square shaped waves and traveling fragments in experiments with the catalytic reaction of $NO$ and $H_2$ on Rh(110) and simulation with a model with nonlinear diffusion, see [211] and [212].

has in fact been used to simulate square shaped waves, see [211], as well as traveling fragments [212]. For an illustration of the latter phenomena, see Fig. 4.1.

Models with nonlinear diffusion usually cannot be treated with analytic methods. Therefore, we constrain the discussion in the next subsection to the case of diffusion of competing species with different anisotropies. However, some general statements regarding the form of propagating nonlinear waves like fronts and pulses in anisotropic media can still be obtained, compare [210] and [211]. The velocities of such waves is dependent on the angular orientation $\phi$ of the normal vector of the front as well as on the local curvature $\kappa$ and described by the function $c(\phi, \kappa)$. If we trace the evolution of the local curvature in an outward propagating wave with $c(\phi, \kappa)$, we may find that for certain angles the curvature increases and sharp corners form [213]. Such corners are expected, if

$$c(\phi, 0) + \frac{\partial^2 c(\phi, 0)}{\partial \phi^2} < 0$$

is fulfilled for some angles $\phi$. Applications of this criterion explains observations made in two-dimensional simulation [213]. For the simple case, discussed below we can derive approximate analytical expression for $c(\phi, \kappa)$ as will be demonstrated in the next subsection.

4.2.2 Anisotropic Pattern Formation in Bistable Media

Note: This section is largely identical with the paper h) listed in the Appendix.

Here we discuss the impact of anisotropy on front propagation in bistable media with
two diffusing species with different anisotropies. Particular emphasis is given to a new dynamical state - stratified spatiotemporal chaos (SSC). A complete treatment of these issues and derivation of the formulas sketched in this subsection can be found in [214]. The model is the FitzHugh-Nagumo model where activator and inhibitor diffusion exhibit linear diffusion with different anisotropies. This model is considerably simpler than equations where nonlinear diffusion causes an effective anisotropy. It allows for an analytic derivation of curvature-velocity relations for different angles. The phenomenon of SSC is demonstrated with numerical simulations and characterized by computing orientation dependent correlation functions. In addition, the mentioned equation for the dependence of interface velocities on parameters, curvature and angular orientation is presented. The mechanism leading to SSC can be described in terms of these analytic results. It is qualitatively different from earlier experimental observations and theoretical studies of spiral chaos in isotropic bistable media [184, 185, 216, 217], where chaotic patterns are due to transitions between counterpropagating fronts induced by curvature or front interactions.

Many qualitative features of pattern formation in chemical and biological reaction-diffusion systems are well described by FitzHugh-Nagumo (FHN) type models for bistable media [218, 219]. The specific model we choose to study is

\[
\begin{align*}
\frac{\partial u}{\partial t} &= \epsilon^{-1}(u-u^3-v) + \delta^{-1}\nabla^2 u + \frac{\partial}{\partial y}[d \delta^{-1} \frac{\partial u}{\partial y}], \\
\frac{\partial v}{\partial t} &= u - a_1 v - a_0 + \nabla^2 v,
\end{align*}
\]

(4.1)

where \(u\) is the activator and \(v\) the inhibitor. The parameters \(a_1\) and \(a_0\) are chosen so that Eqs. (4.1) represent a bistable medium with two stationary and uniform stable states, an “up” state, \((u_+, v_+)\), and a “down” state, \((u_-, v_-)\). Front solutions connect the two states. Pattern formation in these equations has been analyzed by considering the number of front solutions. This number changes, when a single front (an “Ising” front) that exists for values of \(\eta := \sqrt{\epsilon \delta} > \eta_c\) loses stability to a pair of counter-propagating fronts (“Bloch” fronts) for \(\eta \leq \eta_c\). The corresponding bifurcation is referred to as a front bifurcation or nonequilibrium Ising-Bloch (NIB) bifurcation. The anisotropy of the medium is expressed in the parameter \(d\). In general both species, \(u\) and \(v\) have different diffusion constants in the \(x\) and \(y\) direction. In general, the diffusion anisotropy of one of the two species (here the inhibitor) can be removed by a simple rescaling.

We have studied pattern dynamics in Eqs. (4.1) by fixing the value of \(a_1 = 2\) and \(a_0 = -0.1\) (bistable conditions) and varying parameters \(\epsilon\) and \(\delta\) along the line...
4.2 Diusion Anisotropy

Fig. 4.2: Formation of a labyrinth in the isotropic system (upper panel) and formation of stripe in anisotropic system with the same parameter values (lower panel). The thick contour line represents \( u = 0 \) and the thin contour line \( v = 0 \). The shaded regions are domains in the “up” state.

\[ \epsilon = 0.02\delta \]

The anisotropy parameter was changed in a range between 0.5 and 4, most results shown below have been obtained with the choice \( d = 1 \). The following sequence of patterns is observed: at small \( \delta \), stable traveling fronts, pulses and rotating spirals are encountered. For slightly larger \( \delta \) irregular dynamic patterns resembling spiral defect chaos found in the isotropic system [184, 185] appear. Upon further increase of \( \delta \), we find an interesting behavior that has no counterpart in isotropic media, the mentioned state of SSC, that will be described and analyzed in detail in the following.

The existence region of SSC, \( \delta_l(d) < \delta < \delta_u(d) \), increases with the anisotropy parameter \( d \). For the parameters chosen in the simulations, SSC can be found in a region near \( \delta = 1.8 \). For \( \delta > \delta_u(d) \) breathing or stationary periodic stripe patterns are formed parallel to the \( y \) axis. In isotropic media, labyrinths are a typical structure found at large \( \delta \) values, compare Fig. 4.2. The anisotropy suppresses the appearance of these labyrinthine patterns, as is illustrated in Fig. 4.2. SSC is characterized by a general orientation parallel to the \( y \) axis. It is best described as an array of domains that consist of elongated almost stationary pieces of stripes combined with dynamically moving spot-like domains. Fig. 4.3 shows the formation and time evolution of such a
A characteristic property of spatio-temporal chaotic patterns are correlations that decay on a length scale $\xi$ much smaller than the system length $L$. We have computed the normalized spatial two-point correlation functions for the $u$ field both in $x$ and $y$ directions and find that the correlations in the $y$-direction, $C_y(r) = \langle \Delta u(x, y + r) \Delta u(x, y) \rangle / \langle \Delta u(x, y)^2 \rangle$, where $\Delta u(x, y) = u(x, y) - \langle u \rangle$ and brackets $< >$ denote space and time averaging. $C_y(r)$ decays indeed fast to zero, whereas correlations in the $x$-direction, $C_x(r) = \langle \Delta u(x + r, y) \Delta u(x, y) \rangle / \langle \Delta u(x, y)^2 \rangle$, decay for small $r$ to a finite value and then oscillate with constant amplitude (Fig. 4.4). This observation may be used as a definition of the anisotropic state of SSC. It displays finite correlation length in one direction ($x$) and infinite correlation length in the other ($y$).

In isotropic systems, the origin of various complex patterns (labyrinths, spiral chaos)
was explained by considering the front dynamics in dependence on the control parameters $\delta, \epsilon$ and on the local curvature $\kappa$ [184, 185]. The velocity-curvature relations are derived for $\lambda := \sqrt{\epsilon/\delta} \ll 1$. We transform to an orthogonal coordinate system $(r, s)$ that moves with the front, where $r$ is a coordinate normal to the front and $s$ is the arclength. We denote the position vector of the front by $\mathbf{X}(s, t) = (X, Y)$, and define it to coincide with the $u = 0$ contour line. The unit vectors tangent and normal to the front are given by

$$\hat{s} = \cos \theta \hat{x} + \sin \theta \hat{y}, \quad \hat{r} = -\sin \theta \hat{x} + \cos \theta \hat{y},$$

where $\theta(s, t)$ is the angle that $\hat{s}$ makes with the $x$ axis. A point $\mathbf{x} = (x, y)$ in the laboratory frame can be expressed as

$$\mathbf{x} = \mathbf{X}(s, t) + r \hat{r}.$$  \hspace{1cm} (4.2)

This gives the following relation between the laboratory coordinates $(x, y, t)$ and the coordinates $(s, r, \tau)$ in the moving frame:

$$x = X(s, t) - r \sin \theta(s, \tau),$$  
$$y = Y(s, t) + r \cos \theta(s, \tau),$$  
$$t = \tau,$$

where we defined $\mathbf{s} = \partial \mathbf{X}/\partial s$ and $\partial X/\partial s = \cos \theta, \partial Y/\partial s = \sin \theta$. If we express the partial spatial derivatives of $u$ and $v$ after $x$ and $y$ as well as the Laplacians $\Delta u, \Delta v$
in the new coordinates, $s$ and $r$, a new set of coupled partial differential equations is obtained.

Since $\lambda \ll 1$ we distinguish between an inner region where $\partial u / \partial r \sim O(\lambda^{-1})$ and $\partial v / \partial r \sim O(1)$, and outer regions where both $\partial u / \partial r$ and $\partial v / \partial r$ are of order unity. Consider first the inner region. Introducing a stretched normal coordinate $z = r / \lambda$ we obtain at $O(1)$ the stationary front solution

\[ u_0 = -\tanh \left( z \frac{I(\theta)}{\sqrt{2}} \right), \quad v_{f0} = 0, \]

where

\[ I(\theta) = \sqrt{1 + d \cos^2 \theta}. \]

At $O(\lambda)$ we find the equation

\[ C_n = -\frac{3}{\eta \sqrt{2}} I(\theta) v_f - \frac{1 + d}{\delta I(\theta)^2} \kappa. \quad (4.3) \]

Consider now the outer regions to the left and to the right of the front region where $\frac{\partial u}{\partial r} \sim \frac{\partial v}{\partial r} \sim O(1)$. By construction, the outer solution is continuous at $r = 0$. Demanding continuity of the derivative $\frac{\partial v}{\partial r}$ at $r = 0$ as well gives a second relation between $C_n$ and $v_f$. Eliminating $v_f$ by inserting this relation into Eq. (4.3) gives an implicit relation between the normal velocity of the front and its curvature

\[ C_n + \frac{1 + d}{\delta I(\theta)^2} \kappa = \frac{3 I(\theta)(C_n + \kappa)}{\eta \sqrt{2} q^2 \sqrt{(C_n + \kappa)^2 + 4q^2}} + \frac{3 I(\theta) a_0}{\eta \sqrt{2} q^2}, \quad (4.4) \]

where $q^2 = a_1 + 1/2$. A complete account of this derivation is presented in [214]. Relations of this kind can be used to find the NIB bifurcation and predict the stability of planar fronts to transverse perturbations. Also the slopes of the $C_n - \kappa$ relation at nonzero $\kappa$ values indicates the stability or instability to transverse perturbations of curved fronts as well. A multivalued form with branches terminating at realizable curvature values indicates the possibility of spontaneous front transitions. Eq. (4.4) can also be used to study the effects of anisotropy on the NIB bifurcation. Along the line $\epsilon = 0.02 \delta$, the front bifurcation point is for fronts normal to the $x$-direction ($\theta = \pi / 2$) at $\delta_{F,x} \approx 1.382$ for the parameters used here. The anisotropy shifts the bifurcation point for other angles $\theta$ by the factor $1 \leq I(\theta) \leq \sqrt{1 + \delta}$. Consequently, SSC patterns are observed at parameters values $(1.7 < \delta < 1.9)$ for which the fronts in $x$-direction are far into the Ising regime.

In Fig. 4.5a we have plotted the curvature-velocity relation for fronts in the $x$ and $y$ direction with the parameters of the simulation in Fig. 4.3. While the $y$ direction...
is in the Bloch regime close to the front bifurcation, the $x$ direction is far in the Ising regime. In the $x$ direction the single front is moving with negative velocity towards the up state (displayed grey in Figs. 4.3). In the $y$ direction, a fast moving Bloch front with positive velocity usually causes a stretching of the pattern along the $y$ axis. Further insight is obtained from the angle-velocity relation for planar fronts (Fig. 4.5b). One clearly sees the regions of Bloch (multivalued areas) and Ising behavior (single valued areas) along the angular orientation. Another observation from Fig. 4.5b is the increase of the speed Ising front at angles different from $\theta = \pi/2$.

The mechanism for sustaining stratified spatio-temporal chaos is shown in Fig. 4.6. An initial spot is usually transformed into an elongated stripe parallel to the $y$ axis. The stripe initially shrinks, but front interactions eventually cause the inward motion to stop. A circular spot then begins to form at the end of the stripe. This circular spot finally pinches off and the cycle of elongation, spot formation, and pinch-off starts again. The sustained chaotic dynamics stems from a delicate balance between the Ising front speed and the strength of repulsive front interactions in the $x$ direction. As the parameter $\delta$ is increased the front velocity becomes slower. If it is too slow, approaching Ising fronts are repelled and stable stripes form. If it is too fast, the counterpropagating Ising fronts annihilate and the characteristic stripe fragments will not be formed anymore. In the intermediate range pinch-off and formation of circular spots occurs.

Our simulation results above imply that the SSC state is found near a range where breathing stripes form normal to the $x$-direction. Note that for other angles the front speed increases and collapse of counterpropagating fronts results. This explains the pinch-off of the circular droplets. The period in the $x$-direction is determined by the strength of front interactions. This mechanism of chaos generation is distinctly different from the one in an isotropic system. There, chaotic dynamics are caused by spontaneous front transitions near the NIB bifurcation induced by curvature and are directly related to the growth of transverse perturbations. In the anisotropic case the combination of different front dynamics at different angles of propagation is responsible for the occurrence of SSC.

We have demonstrated a new dynamical state, stratified spatiotemporal chaos, in pattern forming anisotropic reaction-diffusion systems. In corresponding isotropic media a sharp boundary between dynamic spiral chaos to stationary labyrinths is found in parameter space, whereas anisotropic media display two distinct transitions. First a spatial periodicity and preferred orientation is introduced into the chaotic dynamics. In
Fig. 4.5: Two views of the relation of Eq. (4.4) corresponding to the parameters of the simulation in Fig. 4.3. (a) The velocity-curvature relation for fronts in the $x$ direction ($\theta = \pi/2$, dashed curve) and $y$ direction ($\theta = 0$, solid curve). (b) The velocity of planar ($\kappa = 0$) fronts at different angles. At $\theta = 0$ there is a pair of stable Bloch fronts and at $\theta = \pm \pi/2$ only a single Ising front exists.
Fig. 4.6: Closeup of the mechanism for stratified spatio-temporal chaos. (a) The tip of a elongated
spot (b)-(c) grows outward. (d) The spot pinches off and (e) propagates away from the tip. (f) The
tip starts growing again and the process repeats. The parameters are the same as in Fig. 4.3.
a second transition, the irregular pattern dynamics settles down and a stationary “ordered” pattern of parallel stripes appears. The two transition points can be identified by measuring the points where the correlations in $x$ and $y$ direction diverge. Stratified spatiotemporal chaos is the newly emerging intermediate state that is expected to play a role in all anisotropic systems with a transition from spatiotemporal chaos to a stationary pattern.
4.3 Heterogeneous Media

4.3.1 Introduction

Spatiotemporal dynamics in reaction-diffusion systems can be altered through the properties (reactivity, diffusivity) of the medium in which they occur. The behavior of chemical concentration fronts in heterogeneous reactive media has been investigated for a variety of geometries in theoretical \cite{220, 221, 222} as well as experimental \cite{223} and computational studies \cite{224}; in excitable media that support spirals and pulses, the influence of small scale heterogeneities on the dynamics of rotating spirals has also been considered \cite{225}. In Rayleigh-Bénard convection, experiments \cite{226, 227} as well as theoretical studies \cite{228} have revealed that modulations of the aspect ratio on a scale comparable to the roll wavelength can lead to changes of the pattern dynamics (drifting rolls and resonance phenomena). Complicated dynamical phenomena have also been reported in studies of striped semiconductor lasers \cite{229}.

Spatiotemporal pattern formation in complicated and composite geometries with active/passive or active/active components is a fascinating subject; a brief representative sampling of experimental and computational observations of catalytic pattern formation during CO oxidation on microdesigned Pt metal crystal surfaces can be found in \cite{233}. When the substrate length scales become comparable to spontaneously arising dynamic length scales (due to reaction-diffusion instabilities), pattern dynamics can be modified or even radically altered, see \cite{83, 236} and a number of related experimental \cite{230, 231, 232, 233} and theoretical studies \cite{224, 233, 234, 235, 236}. An extreme case is given by media with complicated boundaries, where sudden changes in the boundary profile create large curvature perturbation as was demonstrated for a number of systems \cite{237, 238, 239, 240, 83}. The ability to control the geometry and composition of such composites (boundary shape and size, choice of tiling, choice of number and type of components, choice of activity profiles) presents an immensely rich “design parameter” space, that can be utilized not only to explore phenomenology, but to actively influence spatiotemporal dynamics.

At the limit of large heterogeneity “grain size” (compared to the intrinsic length scale of spontaneously arising patterns) the interaction of patterns with inert or active boundaries dominates (e.g. pinning, transmission and boundary breakup of spirals, interaction of pulses with corners, “pacemaker” effects). At the opposite limit of very small or very finely distributed heterogeneity, effective behavior is observed (slight
modulation of pulses, nearly uniform oscillations, effective spirals). Fig. 4.7 shows recent experimental examples for these two limiting cases coming from two different pattern forming chemical reactions. In Fig. 4.7a the catalyst for the reaction has been distributed in triangular subdomains on a length scale somewhat smaller than the width of a single wave. This leads to an effective triangular anisotropy, seen in the form of the outward propagating waves [232]. In Fig. 4.7b a catalytic surface has been microstructured on a scale much larger than the width of a single wave. Various transmission behaviors are observed at boundaries between different media. In this limit direct simulations as in Fig. 4.7c and perturbation theory applied to nonlinear waves such as fronts and pulses are possible strategies for theoretical treatment. For systematical numerical studies, we refer to [233, 236] and [241, 224]. The perturbation theoretical approach for fronts in bistable media is already discussed in [242]. The derivation of front dynamics for various two-component media has been given by Hagberg and Meron [243, 244] and Bode [222]. Extension to pulses (considered as bound states of interacting fronts) and applications to periodically distributed heterogeneities have been considered by Utzny et al. [245, 246]. A rigorous mathematical treatment is found in recent work of Keener et al. [247, 248, 249]. For the opposite limit, it is often possible to replace the model with spatially varying properties by effective equations homogeneous in space. This procedure is called „homogenisation”. An application to reaction-diffusion models is presented in the next subsection.
Fig. 4.7: Experiments and simulations in heterogeneous chemical media. (a) Waves with triangular anisotropy in a Belousov-Zhabotinsky reaction, where the catalysts has been printed only in some parts of the medium (red), after [232]. (b) Patterns in catalytic CO oxidation, inside the letters F, H and I the surface is pure Pt(110) outside one has Pt(110) contaminated with a few percent of Au atoms. Waves hitting planar boundaries show a 1:2 transmission, i.e. every second wave is killed (Experiment by H. H. Rotermund, FHI Berlin). (c) A simulation of transmission behavior of reaction-diffusion waves at boundaries between media with different properties, after [236].
4.3.2 Homogenisation

**Note:** Parts of this subsection are follow closely the thesis work of my Ph. D. student Clemens Utzny (now at Warwick). The results are still unpublished.

This part is concerned with the conditions under which a heterogeneous system is equivalent to a homogeneous system. This equivalence is established on the grounds of significantly different length scales in a heterogeneous reaction-diffusion system.

The method employed in this section is homogenisation. Homogenisation is an important technique that uses the existence of two spatial scales in a system, a microscopic scale and a macroscopic scale. The goal is to achieve equations that describe the system on the macroscopic scale while accounting for influences from the microscopic scale.

An introduction to homogenisation theory in the context of asymptotic methods can be found in [250]. The classical homogenisation theory relies on the separation of two scales. The macroscopic length scale is given by the diffusion length and the microscopic length scale is the characteristic length scale \( L_{het} \) of the heterogeneity. Pattern forming systems with such a property are encountered in biology (intracellular calcium release sites and cardiac tissue) as well as in chemistry (microstructured catalytic surfaces).

We illustrate the homogenisation procedure here for small heterogeneous systems and discuss briefly the generalization for large systems. Small means that the diffusion length of particles is much larger than the system extension, thus allowing the particles to diffuse over the entire system between two reactions. Small heterogeneous systems found in nature are for example cells. They are frequently described by effective equations depending on the spatial extension and the diffusion constants.

Comparable work on homogenisation has been done by Keener [247, 249], who obtains model equations for propagation of electrical signals in cardiac tissue via a homogenisation procedure. Homogenisation of small systems is studied in this section. A heterogeneity \( b(x) \) in a system with length \( L_{sy} \) is shown to yield an effective homogeneous medium described by the average \( < b(x) > \), if the system length \( L_{sy} \) is much shorter than the diffusion length. The following perturbation calculation considers a special form of an reaction-diffusion equation. An extension of the results to more general reaction kinetics is presented at the end of this section. As an example we study the heterogeneous version of the FitzHugh-Nagumo equations:

\[
\partial_t u = f(u) - v + D_u \partial_x^2 u + b_u(x)
\]  

(4.5)
\[ \partial_v = \epsilon(u - av) + D_v \partial_v^2 v + b_v(x). \]  

(4.6)

The upper equations contain the system length \( L_{sy} \) only implicitly via the boundary conditions (either zero flux or periodic boundary conditions). One way of obtaining a representation of Eqs. (4.5, 4.6) that explicitly contain the system length is the Galerkin truncation. Solutions of Eq. (4.6) are represented in the Galerkin truncation by the following ansatz:

\[ u(x, t) = \sum_{n=0}^{\infty} u_n(t) |w_n(x/L_{sy})\rangle \]  

(4.7)

\[ v(x, t) = \sum_{n=0}^{\infty} v_n(t) |w_n(x/L_{sy})\rangle \]  

(4.8)

with \( \langle w_m(x/L_{sy}) | w_n(x/L_{sy}) \rangle = \delta_{mn} \). Inserting this ansatz into the original equations yields after projection on the basis functions \( |w_n(x/L_{sy})\rangle \) a set of coupled ODEs. The fact that the set contains \( L_{sy} \) explicitly allows us to use the small system length \( L_{sy} << 1 \) in a perturbation calculation. The derivation of the ODEs and the perturbation ansatz is presented in the following.

**Perturbation Calculation**

The spatial modes \( |w_n(x/L_{sy})\rangle \) are Fourier modes and thus eigenfunctions of the Laplace operator:

\[ \partial_x^2 |w_n(x/L_{sy})\rangle = -\left( \frac{2\pi n}{L_{sy}} \right)^2 |w_n(x/L_{sy})\rangle. \]  

(4.9)

The boundary conditions determine the specific set of Fourier modes in which solutions of Eq. (4.6) are represented, *i.e.* cosines in the case of zero flux boundary conditions. The particular Fourier modes, however, are not important for the perturbation calculation. The only important property used in the following calculation is the fact that they are eigenfunctions of the Laplace operator. The results are hence independent of the boundary conditions. Using the ansatz of Eq. (4.7) and Eq. (4.8) in the original systems Eq. (4.5) and Eq. (4.6) yields a set of infinitely many coupled ordinary differential equations, each one describing the temporal dynamics of a Fourier mode \( n \). A pair \((u_n, v_n)\) of ODEs is obtained for each mode \( n \). The homogenisation procedure uses these ODEs as the starting point. The general structure of these pairs of ODEs for the modes \( u_n \) and \( v_n \) is given by:

\[ \partial_t u_n = \langle f \left( \sum_i u_i w_i(x/L_{sy}) \right) | w_n(x/L_{sy}) \rangle \]
\[ -D_u \left( \frac{2\pi n}{L_{sy}} \right)^2 u_n - v_n + b_{u_n} \]  

\[ \partial_t v_n = \epsilon (u_n - av_n) - D_v \left( \frac{2\pi n}{L_{sy}} \right)^2 v_n + b_{v_n}. \]  

with \( b_{u_n} = \langle b_u(x) \mid w_n(x/L_{sy}) \rangle \) and \( b_{v_n} \) correspondingly. An appropriate small parameter containing \( L_{sy} \) is given by

\[ \delta = \frac{1}{D_u} \left( \frac{L_{sy}}{2\pi} \right)^2. \]  

This ansatz assume for reasons of notation that \( b_{u_n} \) and \( b_{v_n} \) are order \( O(1) \), a restriction that can be removed by including the terms in the small parameter \( \delta \). The consequences are discussed at the end of this calculation. The results, however, are completely analogous to the ones obtained in the following. Multiplication of (4.10) and (4.11) with \( \epsilon \) results in:

\[ \partial_t u_n = \epsilon \left( f \left( \sum_i \epsilon_i w_i \right) |w_n| u_n - v_n + b_{u_n} \right) - u_n n^2 \]  

\[ \partial_t v_n = \epsilon \left( \epsilon (u_n - av_n) + b_{v_n} \right) - \frac{D_v}{D_u} v_n n^2 \]  

(the explicit spatial dependence of \( w_n(x/L_{sy}) \) is dropped in the following). It should be pointed out that there is only one equation in which \( \delta \) does not occur: the equation for the homogeneous mode \( n = 0 \).

The small parameter \( \delta \) is now used for perturbation analysis. Scaling the time \( t \) with:

\[ \delta \partial_t \rightarrow \partial_r \]  

and expanding each mode \( u_n \) and \( v_n \) in a power series of \( \delta \):

\[ u_n = u_{n,0} + \delta u_{n,1} + \ldots \]  

\[ v_n = v_{n,0} + \delta v_{n,1} + \ldots \]  

gives a hierarchy of equations in \( \delta \) for each mode \( n \), except for the homogeneous mode \( n = 0 \). The equations containing all orders of \( \delta \) are obtained when inserting the upper ansatz into Eq. (4.13) and Eq. (4.14). Collecting terms of the same order yields a simple equation in zeroth order

\[ \partial_r u_{n,0} = -u_{n,0} n^2 \]  

\[ \partial_r v_{n,0} = -\frac{D_v}{D_u} v_{n,0} n^2 \]
with exponentially decaying solutions.

\[ u_{n,0} = c_1 \exp(-n^2 \tau) \]  \hspace{1cm} (4.20)

\[ v_{n,0} = c_2 \exp\left(-n^2 \frac{D_v}{D_u} \tau\right) \]  \hspace{1cm} (4.21)

This result has already some important consequences. First, all modes with \( n \neq 0 \) decouple and decay exponentially fast in the zeroth order of the perturbation calculation \( (L_{sy} \to 0) \). Second, the only ODE with non-decaying behaviour is the one describing the dynamics of the homogenous mode \( u_0 \). The only term that couples this mode with higher ones is \( \langle f(\sum u_i w_i)|w_n\rangle \). Their contribution to the dynamics of \( u_0 \) can be neglected, since only the long time behaviour of the system is under consideration and it reduces to \( f(u_0) \). Hence, the long time behaviour of the system is described in zeroth order by

\[ \partial_t u_0 = f(u_0) - v_0 + b_{u_0} \]  \hspace{1cm} (4.22)

\[ \partial_t v_0 = \epsilon(u_0 - av_0) + b_{v_0} \]  \hspace{1cm} (4.23)

where \( b_{u_0} \) and \( b_{v_0} \) are obtained by integrating over the whole system, \( b_{u_0} = \int b_u(x)dx \) and \( b_{v_0} = \int b_v(x)dx \). The corresponding first order equations are:

\[ \partial_t u_{n,1} = \langle f \left( \sum_i u_{i,0} w_i \right) | w_n \rangle - v_{n,0} + b_{u_n} - u_{n,1} n^2 \]  \hspace{1cm} (4.24)

\[ \partial_t v_{n,1} = \epsilon(u_{n,0} - av_{n,0}) + b_{v_n} - \frac{D_v}{D_u} v_{n,1} n^2 \]  \hspace{1cm} (4.25)

Again only the long time behaviour is considered and the set of ODEs (note the exponentially decaying terms) is given by:

\[ \partial_t u_{n,1} = b_{u_n} - u_{n,1} n^2 \]  \hspace{1cm} (4.26)

\[ \partial_t v_{n,1} = b_{v_n} - \frac{D_v}{D_u} v_{n,1} n^2 \]  \hspace{1cm} (4.27)

The solutions show an exponential decay to a fixed value.

\[ u_{n,1} = q_1 \exp(-n^2 \tau) + b_{u_n} \frac{1}{n^2} \]  \hspace{1cm} (4.28)

\[ v_{n,1} = q_2 \exp\left(-n^2 \frac{D_v}{D_u} \tau\right) + b_{v_n} \frac{1}{n^2} \]  \hspace{1cm} (4.29)

The spatial modes \( |w_n\rangle \) are decoupled in first order of \( \delta \). Though the modes for \( n \neq 0 \) decay exponentially fast they do not necessarily become zero. Their asymptotic value
is determined by the heterogeneity. Hence the dynamics of the solutions is completely determined by the zeroth mode, but spatial variations of this homogeneous mode are induced by non vanishing higher modes.

Altogether, it is found that the temporal behaviour is completely determined by the homogeneous mode whereas the heterogeneity enters in first order. This correction term yields a simple spatial variation but it does not affect the pattern dynamics.

\[
\begin{align*}
\frac{\partial}{\partial t} u_0 &= f(u_0) - v_0 + b_u \\
\frac{\partial}{\partial t} v_0 &= \epsilon(u_0 - av_0) + b_v \\
u_1(x) &= \sum_{n=1}^{\infty} \frac{b_{u_n}}{n^2} w_n(x) \\
v_1(x) &= \sum_{n=1}^{\infty} \frac{b_{v_n}}{n^2} w_n(x)
\end{align*}
\]

(4.30)

Going to second order of $\delta$ is difficult, since the second order terms start changing the actual dynamics of the system, and general statements can not be obtained. The homogenisation results are valid for more complex reaction kinetics $g(u, v)$ and $h(u, v)$, since the linear diffusion operator becomes dominant - the nonlinearities $g$ and $h$ play only a role for the homogenous mode.

Similar behavior is obtained in „long” systems where $L_{sys} >> L_{het}$, where it is however more difficult to cast the perturbation theory results in a general form [245]. However, the equations

\[
\begin{align*}
\frac{\partial}{\partial t} u_0 &= f(u_0) - v_0 + b_u \\
\frac{\partial}{\partial t} v_0 &= \epsilon(u_0 - av_0) + b_v \\
u_1(x) &= \sum_{n=1}^{\infty} \frac{b_{u_n}}{n^2} w_n(x) \\
v_1(x) &= \sum_{n=1}^{\infty} \frac{b_{v_n}}{n^2} w_n(x)
\end{align*}
\]

(4.31)

hold. The first order corrections $u_1(x)$ and $v_1(x)$ are still not time-dependent and do not influence the dynamics of the zeroth order $u_0(x, t)$ and $v_0(x, t)$.

**Applications**

This subsection describes numerical results obtained in systems close to the homogenisation limit. The FitzHugh-Nagumo system is integrated in the oscillatory and in
the bistable regime and the effect of a cosine heterogeneity acting on the activator is studied. The previous computations on homogenisation were fairly general and admitted a simultaneous activator- and inhibitor heterogeneity \((b_u(x)\) and \(b_v(x)\)). This fact required an expansion of both quantities \(\tilde{b}_u\) and \(\tilde{b}_v\) in a power series of \(\delta\). A homogenised description for reaction-diffusion systems where the heterogeneity acts on a non-diffusing component is not possible. Such a heterogeneity is not damped by diffusion, so that it results in large perturbations of the corresponding component.

An extension of the homogenisation procedure to two or more spatial dimensions is straightforward. A checkerboard heterogeneity for example is then given by a function \(b_u(x, y)\) like:

\[
b_u(x, y) = b_{u_n} \cos(nx) \cos(ny).
\] (4.32)

The corresponding Galerkin modes are products of Fourier modes and the appropriate averaged heterogeneities are obtained by integration over both spatial coordinates.

\[
\langle b_u(x, y) \rangle = \int dx \int dyb_u(x, y)
\] (4.33)

The above considerations show that a checkerboard heterogeneity is stronger damped \((\nabla^2 b_u(x, y) = -2n^2b_u(x, y))\) than a stripe heterogeneity \((\nabla^2 b_u(x, y) = -n^2b_u(x, y))\).

\[
b_u(x, y) = b_{u_n}(\cos(nx))
\] (4.34)

This fact is indeed observed in numerical simulations, see also [233].

The influence of periodic heterogeneities on spiral waves in the oscillatory regime as well as on pulses and fronts has been studied in various papers, see [233] and related work [234, 245]. Fig.

Heterogeneities may also be used to direct a pattern forming process. Many processes like dewetting of thin films or spinodal decomposition lead to slowly coarsening labyrinthine patterns. Use of heterogeneities with striped or checkerboard geometry may direct the pattern towards a desired shape. This concept is, for example, used in the fabrication of metal nanoclusters on biotemplates with a periodic structures [257]. This procedure is illustrated in Fig. 4.8, where cluster formation is visible in three out of four frames. However, if no carbon is added to prevent fast diffusion, homogenization sets in and the nanostructured biotemplate has no impact on the pattern formation anymore; we see a typical dewetting pattern. A possible description at the continuum level here is a thin film equation derived from the Navier-Stokes equation [258] which takes into account spatial heterogeneities of the substrate. Again homogenisation would
Fig. 4.8: Experimental snapshots of metal clusters on a biotemplate. The lower right image shows a case, where the deposited Pt is not bound to carbon, it can diffuse freely and destroys the effect of the template.
be a possible strategy, the interesting phenomena however appear if the length scale of the template starts to effect the spatiotemporal dynamics and thus requires direct simulations or other numerical techniques.

### 4.3.3 Conclusion

Diffusion is the basic reason for the homogenisation of reaction-diffusion equations. The diffusion operator ensures smoothness (regularity) of the solutions. Consequently, heterogeneities characterised by a large wavenumber can be treated as small perturbations. This fact is best seen in the Galerkin truncation of these equations. The Galerkin truncation yields a system of coupled ODEs where the heterogeneity $b_u(x)$ with wavenumber $n$ occurs first in the corresponding equation for the $n$th Fourier mode

$$
\partial_t u_n = P_n - D u_n^2 u_n + b_{u_n},
$$

where $P_n$ denotes the projection of an arbitrary nonlinear function. This equations is completely dominated by the diffusion term if the wavenumber $n$ is large. The nonlinear coupling terms can in this case be neglected and thus an effective medium limit is found. The medium behaves linearly in this part of the functional space and thus the first order correction terms result in a superposition principle; they simply yield a spatial modulation of the homogeneous pattern. These results are independent of the specific reaction kinetics and of the number of components in the reaction-diffusion system.

The validity of the homogenised description depends on the diffusion constants $D_u, D_v$, the characteristic heterogeneity length $L_{het}$, the amplitude of the heterogeneity $b_{u_n}, b_{v_n}$ and on the solution itself. A pattern like the homogeneous oscillation is spanned by a few low wave number modes, the dynamics of such a solution is consequently very robust - only heterogeneities of low wave number modes result in significant contributions in the corresponding ODEs. A travelling front is in contrast to homogeneous oscillation described by many Fourier modes and is thus more sensitive to heterogeneities.

This chapter established by means of perturbation calculations that microstructured reaction-diffusion systems show an effective medium behaviour if the characteristic length scale of the heterogeneity is significantly smaller than the diffusion length. The effective equation are simply obtained by adding the spatial average of the heterogeneity to the homogeneous equations, these equations are found in the lowest order of the perturbation calculation. The first order correction terms establish a superposition principle in the homogenisation regime, since they only yield a spatial variation of the
homogeneous pattern.
The characteristic length of the heterogeneity enters quadratically into the expansion parameter whereas the amplitude of the heterogeneity enters linearly, thus showing that the length scale dominates in the homogenisation limit, a fact that explains why even heterogeneities with large amplitudes can be treated as small perturbations in the homogenisation limit.

4.4 Discussion

In this chapter, we studied the formation of complex patterns as a result of anisotropy and heterogeneities. Anisotropy may alter chemical waves and cause sharp corners as well as propagation only along a preferred symmetry axis. In contrast, stationary labyrinthine patterns are typically transformed into aligned stripe patterns. The transition between spatiotemporal chaos and stationary patterns, happens in two steps. One direction already exhibits long range correlation, while the other still shows chaotic dynamics. Overall the new state of stratified spatiotemporal chaos emerges. Many properties of these patterns can be explained and traced back to the angular and curvature dependencies of front resp. pulse velocities. The latter step has been demonstrated in [211] for square shaped waves and in [214] for stratified spatiotemporal chaos.

Heterogeneities introduce a large number of possible effects into pattern formation. A crucial point is the length scale on which the properties of a medium change in a heterogeneous situation. If it is much shorter than the length scale of typical transport processes like diffusion resp. the pattern formation length scale, homogenization procedures can be applied to obtain an effective medium description. In the opposite limit, phenomena like wave reflection and diffraction, formation of spirals at interfaces and pinning of waves to heterogeneities can be expected. In [233], phenomena at the interface of two different media are studied in one and two dimensions. Elimination of waves and formation of spirals at the interface as well as boundary driven spatiotemporal chaos are found. Reference [236] gives an overview of experiments and simulations of catalytic media with inert and active heterogeneities.
5 Perspectives

5.1 Discrete and Stochastic Models

Topics such as stochastic resonance in excitable media [259] wave propagation in noisy, subexcitable media [260, 261, 262, 263] and wave propagation in disordered media [264] have attracted a lot of interest recently. These studies link the fields of pattern formation and stochastic processes and have generated substantial research output [265]. Often, external noise is considered in experiments with random forcing as well as in studies of stochastic partial differential equations. However, microscopic rules may produce a non-negligible amount of internal noise if the number of particles involved in a selforganization process is small (e.g. $10^2 - 10^3$). Two examples for such studies can be found in the appendix of this thesis and are described briefly in the next subsections. On the one hand, we have studied a largely simplified model for the stochastic, discrete dynamics of intracellular calcium channels [252], on the other hand, the formation of rippling patterns in myxobacterial aggregates has been successfully modelled with a discrete, particle-based model that considers the dynamics of individual cells with internal noise [266].

The impact of stochasticity on intracellular calcium dynamics is visible in the experiments by Parker et al. [267] shown in Fig. 5.1. There, waves can be initiated by a local increase in calcium concentration (zap). The outcome of the initial perturbation depends however largely on the number of active calcium channels loaded with the phosphate compound IP$_3$. While a zap leads to an abortive wave that cannot propagate through the medium (Frame A in Fig. 5.1), activation of the calcium channels by photolysis causes visible local fluctuations of calcium release. If several fluctuations towards high calcium concentration appear nearby in space and time, homogeneous nucleation can take place (Frame B in Fig. 5.1). If a zap is applied in an activated
medium, calcium wave initiation is reproducible and reliable (Frames C - F in Fig. 5.1).

The model described in [252] reproduces this behavior and allows for analytical predictions of propagation failure in the limit where discrete effects are dominant over fluctuations. The propagation failure of waves classically described as a pinning bifurcation in deterministic systems [20] should now be classified as a nonequilibrium phase transition [268] where the critical behavior at the onset of propagation is governed by fluctuations. The transition from spark to wave behavior is shown to be in the universality class of directed percolation.

A second example for a discrete stochastic pattern forming system is given by the aggregation phase of myxobacteria [269, 270]. There, standing surface waves known as „rippling” emerge from the initially flat layer of a bacterial aggregate (see Fig. 5.2). Since the discovery of the phenomenon by Reichenbach in 1965 many experimental
studies of rippling have been performed, see [270] and references therein. Unlike many other patterns in bacterial growth and aggregation, rippling has withstood a theoretical explanation until recently [15]. This is in part because continuum reaction-diffusion models cannot capture the essential interaction between the cells, which is reversal upon direct cell-to-cell contact. In [266], it is shown that rippling emerges indeed from the interplay of cell migration and cell-cell collisions [266]. The model has to be discrete, because the wavelength of the pattern is only 200 µm while the individual bacteria have an extension of 10 µm in the direction orthogonal to the ripples. In addition, a refractory time, during which the cells cannot reverse, again has been postulated in the model of Reference [266]. This refractory time determines the wavelength of the standing wave pattern. The wavelength grows monotonously with the refractory time. Furthermore, the columns are only a few cells high. This renders conventional mean-field theory inappropriate. A modification that includes fluctuations around local mean values of densities of right and left traveling cells by a Poisson distribution allows correct predictions of the pattern wavelength found in simulations [271].

Fig. 5.2: Myxobacterial rippling (Reichenbach)
5.2 Control and Forcing

In the previous chapters, we have considered exclusively media with stationary external conditions. Various extensions are possible: First, the periodic properties of heterogeneous media can be replaced by quenched disorder [251]. Second, we can allow for time-dependent heterogeneities (forcing), where the time variation may be either periodic or stochastic. Other successful strategies to alter or control pattern formation include global coupling [272, 273, 274, 275, 276, 277] and resonant forcing of oscillatory systems [8, 278, 279, 280, 281, 282] as well as local [34] and nonlocal [52] feedback strategies. One example, resonant spatiotemporal forcing of oscillatory media near onset is described in [284] showing that depending on the forcing strength, different responses are possible. The response can be harmonic or subharmonic in the wavelength depending on the forcing strength and the involved length scale. Stochastic forcing includes the examples already mentioned in the previous section and maybe useful in improving the propagation of waves in subexcitable media [261] and in realizing ideas about spatial stochastic resonance [259].

5.3 The Future of Pattern Formation

Pattern formation in systems far from equilibrium remains a fascinating and rich subject with many possible paths of development. This thesis has explored two main directions - emergence of complex and chaotic patterns in homogeneous and isotropic systems as well as the impact of symmetry breaking in space and time on spatial self-organisation. The latter point includes issues as diverse as anisotropy, heterogeneities, spatiotemporal forcing and impact of internal noise on biological patterns. Modern day computers allow numerical simulations of realistic models for reaction-diffusion processes in one, two and three dimensions. If it comes to complex patterns, however, more sophisticated strategies are necessary to gain substantial understanding. We have employed continuation algorithms from low dimensional systems and applied them in the study of traveling waves in one spatial dimension and rotating waves in two dimensions. Numerical stability analysis gives insight in the dynamics of unstable structures and explains the often surprising transition from regular to complex patterns. Perturbation theoretical approaches such as equations of motion for interacting coherent structures (fronts, pulses, interfaces) and homogenization procedures have proven to be useful, in particular in heterogeneous and anisotropic systems. Expansion near the
pattern formation transition yield universal amplitude equations (Ginzburg-Landau-equations). These equations are often a good starting point for analysis of more realistic and complicated models such as Navier-Stokes or reaction-diffusion equations.

Many results in this work concern the properties of large amplitude, nonlinear waves in reaction-diffusion systems. Nonlinear waves of this type are known to play a crucial role in biological systems. They are efficient and robust carriers of information and appear prominently for example as electrical signals traveling along neurons. They are also potential candidates for signal transmission and synchronization in multicellular systems, e.g., in the form of intercellular calcium waves. We expect that many of the ideas developed here with simple model systems and verified in chemical reaction will be revealed in experimental study of biological systems.

Finally, biological situations require often the consideration of internal and external noise into the model. Here, we predict that in the future tools from stochastic and statistical physics have to be combined with the approaches from nonlinear dynamics extensively discussed in this thesis. It would be desirable to have a similar variety of methods as the ones presented in this thesis for mostly deterministic systems to characterize stochastic patterns. A representation of microscopic stochastic dynamics in terms of a deterministic and a noise part (stochastic differential equation) would make for a much more efficient analysis and is at the heart of a number of recent activities [285, 286, 287]. In view of the extensive information from experiments, empirical methods have proved very useful to extract the main features of complex patterns. We mention the methods of empirical orthogonal eigenfunctions (Karhunen-Loeve decomposition), time series analysis, fitting of model equations from data as well as numerical stability and continuation algorithms. The methodology and the phenomena described here are not restricted to reaction-diffusion processes in chemical and biological systems, but carry over to applications in hydrodynamics, nonlinear optics and material science. An exciting aspect of future research will be to employ self organization processes on small scales for nanofabrication as well as their use in biotechnological applications.

From a fundamental point of view, the understanding of spatial structuring in living organisms is only at the beginning. It is an open questions to what extent genetics determines biological structures or how much the dynamics of the processes initiated by gene activity plays a role. In the latter stage, models that capture the chemical transformations and transport of involved particles will become crucial. This enterprise can use the knowledge gained in experiments and theoretical studies in pattern forming
systems in the physical sciences.
6 Declaration

1. Hereby, I declare that I wrote this thesis all by myself without illegal means. Passages whose wording or content has been taken from my publications or other sources are marked. I have not attempted a habilitation in another university so far. I have filed for a police document that shall be sent to the Faculty in due time.

2. Hereby I like to explain my contribution to the publications in the appendix of the thesis in order to fulfill the requirement of the „Habilitationsordnung“. The mostly theoretical papers A.1, A.3, A.6, A.7, A. 14 and A.15 have been produced within my group by my Ph. D. students and postdocs, who appear as coauthors. Papers A.2, A.4, A.11 and A.12 have been written jointly with experimental colleagues, my part was usually the model calculations and theoretical interpretations of the presented measurements. Articles A.10 and A.13 are joint work with guest scientist at MPIPKS resp. Prof. Meron’s group in Israel. Finally, the articles A.5, A.8 and A.9 stem from my term as a postdoc in the US and include also cooperations with Diploma and Ph. D. students.
Erklärung


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