Antispiral waves are sources in oscillatory reaction-diffusion media *

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Spiral and antispiral waves are studied numerically in two examples of oscillatory reaction-diffusion media and analytically in the corresponding complex Ginzburg-Landau equation (CGLE). We argue that both these structures are sources of waves in oscillatory media, which are distinguished only by the sign of the phase velocity of the emitted waves. Using known analytical results in the CGLE, we obtain a criterion for the CGLE coefficients that predicts whether antispirals or spirals will occur in the corresponding reaction-diffusion systems. We apply this criterion to the FitzHugh-Nagumo and Brusselator models by deriving the CGLE near the Hopf bifurcations of the respective equations. Numerical simulations of the full reaction-diffusion equations confirm the validity of our simple criterion near the onset of oscillations. They also reveal that antispirals often occur near the onset and turn into spirals further away from it. The transition from antispirals to spirals is characterized by a divergence in the wavelength. A tentative interpretation of recent experimental observations of antispiral waves in the Belousov-Zhabotinsky reaction in a microemulsion is given.

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I. INTRODUCTION

Chemical pattern formation results typically from the interplay of reaction and diffusion and occurs in many reactions in solutions and gels \textsuperscript{1} and in heterogeneous catalysis \textsuperscript{2}. Reaction-diffusion processes are also believed to be at the heart of morphogenesis in biological systems \textsuperscript{3,4}. Rotating spiral waves are probably the most typical structures investigated so far.

They have been found initially in the Belousov-Zhabotinsky reaction \textsuperscript{3}. Since then, they have been frequently observed in a variety of experimental setups including aggregation of slime molds \textsuperscript{5}, catalytic CO oxidation \textsuperscript{6}, cardiac tissue \textsuperscript{7}, and intracellular calcium dynamics in frog eggs \textsuperscript{8}, and glycolytic activity in extracts of yeast cells \textsuperscript{9}.

Spirals that organize the surrounding medium by regular emission of waves are well established under both oscillatory and excitable conditions. If several spirals populate the system then the waves emitted by neighbouring spirals annihilate each other at the boundary of the spirals’ spatial domains. Such boundaries constitute wave sinks. Intuition tell us that these passive objects (where waves arrive and die) do not influence the medium beyond its closest surrounding.

Recently, Vanag and Epstein reported on experiments of the oscillatory Belousov-Zhabotinsky reaction in an Aerosol OT microemulsion (BZ-AOT) \textsuperscript{10}. In their quasi-two-dimensional system chemical concentration patterns are arranged as so-called “antispirals” or “antitargets”. Typical snapshots of the system display closely packed domains each containing a single antispiral. As time progresses the concentration waves propagate towards the center of the respective antispiral. Such inward propagation seemingly contradicts our intuition of spiral cores as wave sources. A word of caution towards the terminology of spirals and antispirals is in place here: Spirals and antispirals are not structures that can appear simultaneously for given parameters. Instead, they occur in different complementary regions of the parameter space. The terminology of antispirals used here is rather recent and occurs only after the cited experimental paper, that is more careful and talks of inwardly rotating spiral waves. It should also be stressed that many authors have found (what is now often called) antispirals or antitargets in numerical simulations in continuous and discrete oscillatory and chaotic media and simply classified them as spirals or targets see e.g. \textsuperscript{11,12,13}. More recent reports have adapted the new name antispiral in the sense it is used here \textsuperscript{11,13}. The distinction achieved here by the prefix anti is sometimes also referred to as positive dispersion (spiral) and negative dispersion (antispiral) \textsuperscript{11,13}.

Since, the phenomena of antispirals and antitargets occur frequently in the complex Ginzburg-Landau equation \textsuperscript{14}, they should be generic in oscillatory media near onset. What comes as a surprise is rather that over so many years only spirals have been observed and the first experimental report of an antispiral occurred only very recently \textsuperscript{11}. This experiment lead to a discussion about the “mechanism” of antispirals. Vanag and Epstein sug-

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gested that the phenomenon may be related to a Hopf bifurcation with finite wavenumber (wave bifurcation) in a extended Oregonator model, where they find group and phase velocities with opposite signs. Simulations and experiments show also “packet waves”, which are wave groups that move in opposite direction to the motion of the constituting waves [22]. While this argument seems valid for the quasi one-dimensional packet waves, it does, in our opinion, not apply to the two-dimensional case under consideration. After all, near a wave bifurcation in a two-dimensional medium, one typically observes traveling or standing stripes or hexagons and not spiral waves. Earlier on, Nicola et al. have reported related wave groups near a Turing-wave bifurcation and named them “drifting pattern domains” [23].

Gong and Christini have recently investigated the CGLE and prototypical two-component reaction-diffusion (RD) models and conjectured that antispirals only occur near the onset of oscillations [24]. We will provide further evidence for their claim. They also suggest an analytical argument and criterion for the appearance of antispirals in the CGLE coefficients. Recently, we have shown that the criterion for the occurrence of antispirals in RD models has to be modified and that antispirals in the corresponding CGLE may turn out to be spirals in the original RD model [25]. Here we present the complete analytical derivation of the differing criteria for antispirals in the CGLE and in the corresponding RD models.

When studying dynamical phenomena in RD models near the onset of pattern formation then the analysis of the corresponding amplitude equation can be instructive [20, 21, 22]. The CGLE represents the amplitude equation for pattern dynamics near the onset of oscillations via supercritical Hopf bifurcations [21]. The CGLE for arbitrary RD systems is obtained following exact transformation rules [12, 28]. Here, we validate our criterion for antispirals in RD systems for two simple reaction-diffusion (RD) systems: the FitzHugh-Nagumo and Brusselator models by deriving the CGLE coefficients for these models, applying our criterion for antispirals and comparing the result to direct numerical simulations near a Hopf bifurcation. In the CGLE, domains of the parameter space can be classified according to the relative directions (signs) of phase- and group-velocities in rotating waves (spirals or antispirals) in line with arguments which Y. Kuramoto and T. Tsuzuki used first to analyse wave sources of the Kuramoto-Sivashinsky equation [31]. Boundaries of the parameter domains are given by zero phase or group velocities which yields analytical criteria. Extensive numerical simulations of both RD models analysed here, corroborate this criterion’s prediction near the onset of oscillations and extend it when the model’s parameters are driven away from threshold. The simulations also suggest that antispirals typically disappear far away from the onset of oscillations.

This paper is organised as follows: In the next section we will discuss theoretically the distinction between spirals and antispirals within the CGLE framework and analytically derive the criterion for either occurrence in RD models near the onset of oscillations. In the third section we explore spirals and antispirals in two RD models near and far from the onset of oscillations. We will end this paper with a short summary and discussion of the main results.

II. WEAKLY NONLINEAR THEORY OF SPIRALS AND ANTISPIRALS

A. RD systems near a oscillatory bifurcation

A general reaction-diffusion system in two dimensions may be described by the partial differential equation

\[ \partial_t u = f(u, \mu) + D \nabla^2 u, \]  

(1)

where \( u(x, t) \) is a vector of space- and time-dependent concentrations, \( f \) is a nonlinear vectorial function, \( D \) is a diffusion tensor and \( \mu \) is a control parameter. Realistic models of chemical pattern forming systems have been proposed in this form including recent models of the BZ-AOT system by Vanag et al. [22, 31] and Yang et al. [32]. These include many reaction species, respectively components of \( u \), and complex reaction terms \( f \). However, in this paper we shall study simple models namely the Brusselator [32] or the FitzHugh-Nagumo model [33]. (Anti)spiral waves in the latter two models will be studied in Sec. III and explained by results of the weakly nonlinear analysis that we carry out in the present section.

Near the supercritical onset of homogeneous oscillations in the RD system with frequency \( \Omega \) and eigenvector \( u_1 \) the vector of concentrations \( u \) may be decomposed as

\[ u(x, t) = u_0 + u_1 \tilde{A}(x, t) e^{i \Omega t} + u_1^* \tilde{A}^*(x, t) e^{-i \Omega t}. \]  

(2)

The evolution of modulations \( \tilde{A}(x, t) = \sqrt{\epsilon} A(x, t) e^{ic_0 t} \) of a homogeneous oscillation is described by the CGLE [27]

\[ \partial_t A = A + (1 + ic_1) \Delta A - (1 - ic_3) |A|^2 A. \]  

(3)

Here \( \epsilon = (\mu - \mu_c)/\mu_c \) measures the distance from the threshold \( \mu_c \), \( c_1, c_3 \) give the linear (nonlinear) dispersion and \( c_0 \) an overall linear frequency shift. The coordinates \( x = \sqrt{\xi} \) and \( t = \tau \) of the CGLE are defined by characteristic spatial and temporal scales \( \xi, \tau \) and describe slow modulations in space and time. Note, that intrinsic CGLE frequencies \( \omega \) result only in a small correction of order \( \epsilon^\omega \) to the original frequency of the system at the Hopf bifurcation threshold. In the following, variables with (without) tilde will be used in the original RD system (the derived CGLE).

Numerical simulations of the CGLE in a two-dimensional system with zero flux boundary conditions provide examples of antispiral waves as shown in Fig. 1.
These (anti)spiral waves of the two-dimensional CGLE have the form \[ A(r, \theta, t) = F(r)e^{i(\sigma \theta + f(r, t))}, \] in polar coordinates \((r, \theta)\). The asymptotic behaviour for \(r \to 0\) is \(\partial f(r, t)/\partial r \sim r, F(r) \sim r\) and denotes a topological defect \(|A| = 0\) of charge \(\sigma\) in the spiral core. We will focus on one-armed spiral waves with \(\sigma = +1(-1)\) which are related by mirror symmetry and rotate clockwise (counterclockwise). Other authors term these solutions spiral and antisprial \[ \text{[graphic]} \] (we will not adopt this terminology here). In this paper, both left-handed or right-handed structures can be spirals (outward propagation of phase waves) or antispirls (inward propagation of phase waves) depending on parameters.

Asymptotically for \(r \to \infty\) these phase waves are characterised by \(F(r) \sim \sqrt{1 - q_s^2}\) and \(f(r, t) \sim q_s r - \omega_s(q_s) t\) with a selected wavenumber \(q_s\) and the corresponding frequency \(\omega_s(q_s) = -c_3 + q_s^2(c_1 + c_3)\). The selected wavenumber \(q_s\) is a function of the parameters \(c_1\) and \(c_3\) given by the nonlinear eigenvalue problem resulting form inserting Eq. (4) into the CGLE \[ \text{[graphic]} \].

The asymptotic wave field \(f(r, t)\) may be rewritten as \(q_s(r - v_{ph}t)\) with the phase velocity \(v_{ph} = \omega_s/q_s\). The propagation of small perturbations of the wave is described by the group velocity \(v_{gr} = \partial \omega_s/\partial q_s = 2q_s(c_1 + c_3)\). For positive group velocity the spiral or antisprial acts as a source that organises the surrounding pattern. Phase and group velocity do not necessarily point into the same direction. The phase waves move outward (inward) for positive (negative) phase velocity \(v_{ph}\) in the CGLE coordinates or \(\tilde{v}_{ph}\) in the RD system, respectively. Hence, the signs of the selected group and phase velocities constitute the defining quantities for the occurrence of spirals or antispirls.

In the following we calculate the parameter dependence of the introduced velocities and focus on their signs. The selected wavenumber \(q_s(c_1, c_3)\) needs to be computed numerically in general, only its asymptotic are known analytically \[ \text{[graphic]} \]. However, for the one-dimensional (1D) analog of the (anti)spiral wave \(i.e.\) for the 1D CGLE

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**FIG. 1:** Numerical simulation of the CGLE \[ \text{[graphic]} \] revealing antispirals and antitargets for \(c_1 = 1, c_3 = 0.5\). In (a) the real part of \(A\), in a system of size 250*250 with zero flux boundary conditions and after a transient of \(t = 125\), is shown. White (black) areas correspond to maximum (minimum) values. The black arrows indicate the direction of propagation of phase waves and the white arrow denotes the rotation of an individual spiral. The inwardly propagating target waves are induced by a small oscillating heterogeneity in their center. The domain boundaries, where new waves emerge and split, are visible between the antispirals. In (b) a space-time plot of \(\text{Re}[A]\), along a horizontal cut at \(y = 60\) (this line contains the cores of the antispirals at \(x = 75\) and \(175\), is shown. After a transient of \(t = 125\) the final state leads to the configuration shown in (a). Initially the cores are placed in the homogeneously oscillating background. This homogeneous background is suppressed as the antispirals grow until the whole system is filled by the antispirals at \(t = 60\). Note how new waves emerge and split at the domain boundary at \(x = 125\). The evolution of a small perturbation in (b) illustrates the different sign of the phase and group velocities \(v_{ph}\) and \(v_{gr}\) (the artificial perturbation is applied at \(x = 90, t = 65\) and propagates with \(v_{gr}\)). The space-time plot shown in (c) corresponds to the same situation than in (b) but a fast homogeneous oscillation with \(\Omega = 0\) has been added to the phase of \(A\). This illustrates the dynamics in the underlying RD system which, for the value of \(\Omega\) chosen, also shows antispirals \(i.e.\) the phase velocity \(\tilde{v}_{ph}\) and the group velocity \(\tilde{v}_{gr}\) have the same direction as in the CGLE.
in $[0, \infty)$ with Dirichlet boundary condition: $A(0, t) = 0$) the corresponding selected wavenumber $q_{s1}(c_1, c_3)$ is known analytically \([35, 37]\):

\[
q_{s1} = -\frac{3\alpha(c_1, c_3) + \sqrt{\frac{9\alpha(c_1, c_3)^2 + c_3 + 2c_1\alpha(c_1, c_3)^2}{c_1 + c_3}}}{2(c_1 + c_3)},
\]

where $\alpha(c_1, c_3) = \sqrt{3c_1(8(c_1 - c_3)^2 + 9(1 + c_1c_3)^2 - 4c_1c_3)^{1/2} + c_1(5 - 9c_1c_3) - 4c_3}/(4(-2c_3 + 9c_1^2 + 7c_1))$. \(5b\)

The function $\alpha(c_1, c_3)$ is symmetric and $q_{s1}(c_1, c_3)$ antisymmetric under the substitution $(c_1 \rightarrow -c_1, c_3 \rightarrow -c_3)$. The same property holds for the two-dimensional (anti)spirals as can be deduced from the form of the nonlinear eigenvalue problem \([35]\):

\[
q_s(c_1, c_3) = -q_s(-c_1, -c_3), \quad (6a)
\]
\[
q_s(c_1, c_3) = 0, \quad \text{for } c_1 + c_3 = 0, \quad (6b)
\]
\[
q_s(c_1, c_3) > 0, \quad \text{for } c_1 + c_3 > 0, \quad (6c)
\]
\[
q_s(c_1, c_3) < 0, \quad \text{for } c_1 + c_3 < 0. \quad (6d)
\]

Hence, $q_s$ takes the same sign as $c_1 + c_3$. The corresponding frequency $\omega_s(c_1, c_3)$ in the CGLE fulfills $\omega_s(c_1, c_3) = -\omega_s(-c_1, -c_3)$ as had already been noted by Paullet et al. \(35\).

The curve where the selected frequency $\omega_s$ vanishes has been computed numerically in the $(c_1, c_3)$ parameter space (i.e., we computed the parameters $c_1$ and $c_3$ for which $\omega_s(q_{s1}(c_1, c_3)) = 0$). It turns out to be different from the diagonal $c_1 + c_3 = 0$ where $q_s = 0$. Hence, within the CGLE the parameter space, we can distinguish four different domains, as shown in Fig. 2. When crossing the boundaries between these domains, the phase velocity $v_{ph}$ in the CGLE switches sign outward and inward. Note, that the spiral wave length and $v_{ph}$ diverge on the diagonal and $v_{ph}$ crosses zero where $\omega_s = 0$. In the coordinate frame of the CGLE, there are two domains with spirals and two with antisolitons. This result is in agreement with that of Gong et al. \(24\) and additionally covers two quadrants previously not considered, where the transition curve $v_{ph} = 0$ lies. Inserting Eq. \(6a\) into $v_{gr} = 2q_s(c_1 + c_3)$ one finds that the group velocities $v_{gr}$ of both spirals and antisolitons never become negative.

![FIG. 2: Parameter space $(c_1, c_3)$ of the complex Ginzburg-Landau equation (CGLE) and domains where spiral or antisolitonal solutions exist in the corresponding RD systems (where we assumed that $\Omega > 0$). The group velocity of spirals and antisolitons is non-negative everywhere. Note in the CGLE the phase velocity vanishes along the dashed curve and antisolitons (spirals) occur in the upper (lower) shaded and lower (upper) white regions. The dot-dashed curves denote the Benjamin-Feir instability $(1 + c_1c_3 = 0)$. The white circle indicates the location of parameter values used in Fig. 1.](image)

### C. Spirals and antisolitons in RD systems

Now we return to the initial RD system. Therein the asymptotic concentration waves $u \sim e^{iq_s(F - \tilde{v}_{ph}t)}$ with wavenumber $\tilde{q}_s = \sqrt{\tilde{v}_{ph}/\xi}$ read, after inserting Eq. \(1\) into Eq. \(2\),

\[
\tilde{v}_{ph} = \left[-\Omega + \frac{\xi}{\tau}(\omega_s - c_0)\right]/\tilde{q}_s.
\]
The description given by the CGLE \(^3\) is accurate only if \(\epsilon \ll 1\). In this limit \(\tilde{v}_{ph}\) may be simplified as 
\[
\tilde{v}_{ph} \approx -\Omega/q_s \sim -\Omega/q_s \quad \text{(note that the same result follows for the complex conjugate (c.c.) in Eq. \(\text{4}\), where two negative signs cancel)}.
\] 
For the group velocity we find 
\[
\tilde{v}_{gr} = v_{gr} \sqrt{\gamma} \sim v_{gr} \quad \text{which does never get negative.}
\] 
Therefore, both spirals and antispars will act as organising centers of the surrounding concentration pattern.

In order to determine the parameter dependence of the phase velocity \(\tilde{v}_{ph} \sim -\Omega/q_s\), we first assume \(\Omega > 0\). Then spirals with positive phase velocity occur in RD systems with corresponding \(c_1 + c_3 < 0\) as follows from Eq. \(\text{6c}\). Antispars have \(q_s > 0\) and arise in RD systems for which \(c_1 + c_3 > 0\). Consequently, spirals and antispars can never occur simultaneously within a single homogeneous system.

These analytical results are summarised in Fig. \(\text{2}\) (where \(\Omega > 0\) was chosen).

Let us now briefly comment on the case of \(\Omega < 0\) (note that the remarks that follow are not important since by convention a \(\Omega > 0\) is considered in order to calculate the amplitude equation \(\text{4}\)). Assuming the opposite sign of the primary oscillation \(\Omega \rightarrow -\Omega\) yields the same results for the group and phase velocities. For negative \(\Omega\) we find \(c_1 \rightarrow -c_1\), \(c_3 \rightarrow -c_3\), \(\omega \rightarrow -\omega\) as follows from complex conjugation of Eq. \(\text{3}\) and \(q_s \rightarrow -q_s\) as in Eq. \(\text{6c}\).

This gives the condition \(c_1 + c_3 < 0\) for the existence of antispars if \(\Omega < 0\).

Using Eqs. \(\text{8}\) and \(\text{6c}\), we compare the temporal frequencies \(f = -q_s \tilde{v}_{ph}\) measured for a spiral \((f_S)\) or antispiral \((f_{AS})\) wave with that of the homogeneous oscillation \(f_{bulk}\)

\[
\begin{align*}
 f_{bulk} &= \Omega + \frac{\epsilon}{\tau} (c_0 + c_3) , \\
 f_S &= \Omega + \frac{\epsilon}{\tau} (c_0 + c_3) + \frac{\epsilon}{\tau} q_s^2 |c_1 + c_3| , \\
 f_{AS} &= \Omega + \frac{\epsilon}{\tau} (c_0 + c_3) - \frac{\epsilon}{\tau} q_s^2 |c_1 + c_3| .
\end{align*}
\]

We conclude \(f_S > f_{bulk}\) and \(f_{AS} < f_{bulk}\) in agreement with experimental observations \(\text{11}\).

Let us finish this section with a short summary. The observed antispiral waves with phase velocity pointing inward were shown to be organising centers since their group velocity points outward. Antispars in oscillatory media therefore determine the surrounding pattern as it is the case for spiral waves. We found that a spiral in the CGLE coordinate frame may represent an antispiral in the original RD system and vice versa (see Fig. \(\text{2}\)). Altogether, in RD systems near the onset of oscillations, the antispars are predicted to occur if the corresponding CGLE coefficients fulfil \(c_1 + c_3 > 0\).

### III. REACTION-DIFFUSION MODELS

In order to test the general predictions of the previous section we will now address our attention to a specific RD systems. The RD systems of activator-inhibitor type are defined by \(\text{28}\)

\[
\begin{align*}
 \partial_t u &= f(u,v) + \nabla^2 u , \\
 \partial_t v &= g(u,v) + \delta \nabla^2 v ,
\end{align*}
\]

where \(\delta\) is the ratio of diffusion constants and the functions \(f(u,v)\) and \(g(u,v)\) define the dynamics of the activator \(u(x,t)\) and inhibitor \(v(x,t)\), respectively.

In the following we will consider two prototypical examples of activator-inhibitor dynamics widely analysed in the literature: the FitzHugh-Nagumo and Brusselator models. In both cases we will proceed as follows: First we will study their fixed points and analyse their linear stability. The next step will be the derivation of the coefficients \(c_1\) and \(c_3\) of the CGLE \(\text{3}\). They can be used to decide the question whether spirals or antispars appear near the Hopf bifurcation. Finally we will compare these predictions with numerical simulations of both models near and far from onset.

#### A. FitzHugh-Nagumo model

The FitzHugh-Nagumo (FHN) dynamics \(\text{31}\) is defined by \(\text{31}\)

\[
\begin{align*}
 f(u,v) &= u - \frac{u^3}{3} - v , \\
 g(u,v) &= \varepsilon (u - \gamma v + \beta) ,
\end{align*}
\]

where \(\varepsilon > 0\) is the ratio between the time scales of both fields, and \(\beta\) and \(\gamma\) are parameters that determine the number of fixed points. The coordinates of these fixed points are independent of \(\varepsilon\) and are given by the roots of a cubic polynomial. In the following we will only consider \(\gamma > 0\) and restrict our analysis to the case where a unique fixed point \((u_0, v_0)\) exists. This is the case, for example, for \(\gamma = \frac{1}{2}\) where the coordinates of the fixed point are

\[
\begin{align*}
 u_0 &= ((\sqrt{1 + 9\beta^2} - 3\beta)^\frac{3}{2} - 1)/((\sqrt{1 + 9\beta^2} - 3\beta)^\frac{3}{2} + 1)\quad \text{and} \\
 v_0 &= (u_0 + \beta)/\gamma.
\end{align*}
\]

Choosing \(\varepsilon\) as the control parameter and keeping \(\beta\), \(\delta\) and \(\gamma\) constant, a linear stability analysis of \((u_0, v_0)\) shows that it is unstable to periodic oscillations if \(\varepsilon < \varepsilon_H^\star\) where \(\varepsilon_H^\star = (1 - u_0^2)/\gamma\). This Hopf bifurcation has a frequency \(\Omega = \sqrt{(1 - u_0^2)/\gamma - (1 - u_0^2)^2}\) at onset. The fixed point may also become unstable to spatially periodic perturbations with wavevector \(\tilde{q}_T\) defined by \((2\pi)/(\sqrt{1 - u_0^2} + 2\gamma(1 - u_0^2)/\gamma)\) (i.e. a Turing instability) if \(\varepsilon < \varepsilon_T^\star\), where \(\varepsilon_T^\star = ((2 - 1 - u_0^2)\gamma + 2\sqrt{1 - (1 - u_0^2)\gamma})^\gamma/\gamma^2\).

These two instabilities may occur simultaneously at a codimension-2 instability. The thick full line in Fig. \(\text{8a}\) shows the location of this codimension-2 instability in the parameter space \((\beta, \sqrt{\delta})\) for \(\gamma = \frac{1}{2}\). In the following we will restrict our analysis to the Hopf instability (i.e. the region below the codimension-2 line in Fig. \(\text{9a}\)).
instability is predicted. The thin dotted lines indicate the range of parameter values considered in the numerical simulations. In (a) we consider the parameter space $\beta$, $\gamma u^2$, and $1 + c$ transition to antispirals are predicted to exist near the Hopf instability threshold for the FHN and Brusselator models (in (a) and (b), respectively). The thick full lines separates the regions where either the Turing or Hopf instability appears first as the control parameter $\varepsilon$ (parameter $b$) is decreased (increased) away from threshold for the FHN model in (a) (respectively, in (b) the Brusselator model). Inside the dot-dashed lines the Benjamin-Feir instability is predicted. The thin dotted lines indicate the range of parameter values considered in the numerical simulations of (anti)spirals summarised in Figs. 3 and 4. The Hopf bifurcation considered in both figures is supercritical (see main text). In (a) we consider the parameter space $(\beta, \sqrt{\delta})$ of the FHN model (Eqs. 11) with $\gamma = \frac{1}{2}$. For $\beta \geq 2/3$ the fixed point $(u_0, v_0)$ is stable and if $\varepsilon$ is small enough, excitability is possible. In (b) the control parameter $(a, \sqrt{\delta})$ of Eq. 12 is plotted.

will only quote the main results of this derivation. The linear dispersion coefficient is given by

$$c_1 = \left(1 - \frac{u_0^2}{\Omega^2}\right) \frac{\delta - 1}{\delta + 1}.$$ 

Note that this coefficient vanishes if both fields diffuse with equal strength. If $(1 - \gamma - \gamma u_0^2)/(1 - \gamma + \gamma u_0^2) > 0$ the Hopf bifurcation is supercritical, otherwise it is subcritical and the CGLE (3) can not be applied. For $\gamma = \frac{1}{2}$ (the case shown in Fig. 3(a)) the Hopf instability is supercritical for any value of $\beta$. The result for the nonlinear dispersion coefficient $c_3$ is more complicated:

$$c_3 = \frac{3 - 3\gamma - 7u_0^2 + 3\gamma u_0^4}{3 - 3\gamma - 3\gamma u_0^2} \sqrt{\frac{\gamma}{(1 - u_0^2)(1 - \gamma + \gamma u_0^2)}}.$$ 

We may now use the expression for the coefficients $c_1$ and $c_3$ to evaluate the conditions for the spiral-antispiral transition $c_1 + c_3 = 0$ and the Benjamin-Feir instability $1 + c_1c_3 = 0$ in the parameter space $(\beta, \sqrt{\delta})$. These two lines are plotted in Fig. 3(a). Note that the region where antispirals are predicted to occur near threshold, exists only for small values of $\beta$ (i.e. far from the excitability region) and vanishes if $\delta$ is too big or if $\delta \to 0$. Moreover, for $\beta = 0$, where the model exhibits the symmetry $(u,v) \to (-u,-v)$, only antispirals are predicted.

The predicted spiral-antispiral transition at $c_1 + c_3 = 0$ is strictly valid only close to supercritical Hopf bifurcations (i.e. for $\varepsilon \lesssim \varepsilon_{\mu b}$). In order to test this and in addition to investigate the behaviour far from threshold, we have studied the FHN model numerically. We have done extensive numerical simulations of 1D-(anti)spiral analogues in Eq. 3 near and far from threshold. These simulations were performed in very long systems (typically including hundreds of spiral wavelengths) with the following boundary conditions: in the right side a zero flux condition is imposed and in the left side the field is kept to $u = u_0$ and $v = v_0$. The output of these numerical simulations, see Fig. 4, is insensitive to initial conditions. If we wait long enough, the boundary condition in the left side (the "core") will select phase waves with a particular $\tilde{q}_{\mu 1}$ which eventually invade the rest of the system (since they always have positive $\tilde{v}_{\mu 1}$). Antispirals (spirals) will have negative (positive) phase velocity $\tilde{v}_{ph}$. Near the Hopf threshold the observed behaviour is equivalent to the one predicted by the CGLE. But, as $\varepsilon$ is decreased, a transition from antispirals to spirals is seen. This transition is related to a change in the sign of $\tilde{q}_{\mu 1}$ and is not captured by the CGLE description (see below).

In order to assess the previous results for the 1D-(anti)spiral analog we have also carried out a number of 2D simulations of the FHN model near onset. The outcome of these simulations is shown in Fig. 4(b) and confirms that, at least near the onset, the 1D analog provides a good description of the spiral-antispiral transition mechanism in the 2D system. To see this more quantitatively, in Fig. 5 we plot, against $\varepsilon$, the selected wavenumbers $\tilde{q}_s$, $\tilde{q}_{\mu 1}$, in 1D and 2D, and the correspond-
Let us finally address the Brusselator dynamics [33]. This model is defined by:

\[ f(u, v) = a - (b + 1)u + u^2v, \quad (12a) \]
\[ g(u, v) = bu - u^2v, \quad (12b) \]

where \(a\) and \(b\) are two parameters (assumed in the following to be positive). This model has a unique fixed point \((u_0, v_0) = (a, b/a)\), for any value of the parameters \(a, b\) and \(\delta\).

Similarly to the FitzHugh-Nagumo model, we may now examine the stability of the fixed point. Let us take \(b\) as the control parameter. Depending on the values of the parameters \(a\) and \(\delta\), the fixed point \((u_0, v_0)\) may become unstable in two different ways. If \(b > b_H^c\), where \(b_H^c = 1 + a^2\), then a Hopf instability with frequency \(\Omega = a\) occurs.

In the other hand, if \(b > b_T^c\), where \(b_T^c = (1 + a/\sqrt{\delta})^2\), a Turing instability with wavenumber \(q_T^c = a/\sqrt{\delta}\) takes place. These instabilities may occur simultaneously in a codimension-2 line. This line and the regions in the parameter space \((a, \sqrt{\delta})\) where either the Hopf or Turing instabilities appear first, as the control parameter \(b\) is increased, are shown in Fig. 3(b).

A short calculation [26] shows that the coefficients \(c_1\) and \(c_3\) are

\[ c_1 = a \frac{\delta - 1}{\delta + 1} \quad \text{and} \quad c_3 = \frac{7a^2 - 4 - 4a^4}{3a(2 + a^2)}. \]

The spiral/antissipral transition line \(c_1 + c_3 = 0\) is plotted in the parameter space \((a, \sqrt{\delta})\) in Fig. 3(b). Note that the region where antissiprals are predicted is rather small and is located near the area where of the Turing instability appears first. Also in this case the antissiprals will occur only for intermediate values of the diffusion ratio \(\delta\).

We have performed numerical simulations also for this model. As for the FHN model, we investigated 1D-spirals and antissiprals (with the same boundary conditions as in the previous study). The results of these simulations are resumed in Fig. 6. In this figure we also show the output of some 2D simulations, which confirm the observation that 1D simulations provide a good estimate for the location of the spiral-antissipral transition. Also here it is seen that the prediction based in the CGLE description is valid only in the vicinity of the Hopf threshold. As
FIG. 5: Plot of the measured wavenumber \( \tilde{q}_s \) selected by 1D- and 2D-(anti)spirals (open circles and squares, respectively) in numerical simulations of the FHN model as a function of the parameter \( \varepsilon \), with \( \sqrt{\delta} = 1 \) and \( \beta = 0 \) (in this case \( \gamma = 1/2 \) and \( \varepsilon_H = 2 \)). The full lines indicate the wavenumber selected by the 2D antispirals and the 1D antispiral analogues in the CGLE. The CGLE 1D line is calculated using the wavenumber \( \tilde{q}_s^1 \) predicted analytically by Hagan's formula. In the inset the value of \( \tilde{q}_s^2 \) in the region close to the onset is shown. Note that the CGLE 1D and CGLE 2D lines are directly proportional to the distance to the threshold \( \sqrt{\varepsilon_H} - \varepsilon \) and consequently that both \( q_s^1 \) and \( q_s \) are constant in the framework of the amplitude equations, but also note that this constant value differs. The measured \( \tilde{q}_s^1 \) and \( \tilde{q}_s \) approach (as expected) the predictions of the 1D and 2D CGLE antispirals near the threshold but are systematically smaller far away from it.

the distance to this threshold increases the antispirals are turned into spirals.

IV. SUMMARY

In this paper we emphasise the fact that the essential difference between spirals and antispirals is the sign of phase velocity of the travelling waves emanating from the core region and that phase and group velocity do not necessarily have to point into the same direction. Consequently, we may conclude that spirals and antispirals can not occur simultaneously. They appear in different regions of the parameter-space.

We have shown that spirals and antispirals are related by a simple symmetry transformation in the parameter space of the CGLE and derived a criterion for the CGLE coefficients, namely \( c_1 + c_3 > 0 \), that predicts the existence of antispirals in the respective RD model near a Hopf instability. In the CGLE parameter space, antispirals appears as frequently as spirals. At the boundary between antispirals and spirals, the selected wavenumber is zero and the selected wavelength diverges.

Our criterion has been mapped to the parameter space of two representative reaction-diffusion models. The analysis of both models suggests that to get antispirals it is necessary that the inhibitor also diffuses. Moreover, the diffusion constants of both reactants should be of similar magnitude in order to find antispirals, which is likely to occur near the codimension-2 Turing-Hopf instability. We have also performed numerical simulations of both models near and far form the oscillatory instability threshold. Antispirals are quite likely to appear near the Hopf bifurcation but turn into regular spirals far into the oscillatory where the amplitude of waves becomes large.

Both examples show that the assumption that the 1D spiral analog gives a good approximation of the spiral-antispiral transition behaviour not only near onset but also moderately far from it.

In a more general distinction, antispirals emit phase waves that are typical for oscillatory media. In contrast, trigger waves common in excitable media should always have the same sign of group and phase velocity. Since the distinction between trigger and phase waves is not sharp, we cannot make a general statement about the
possibility of ant spirals in excitable media, but expect that they will be quite hard to find therein. The rarity of experimental observations of ant spirals suggests that most experiments exhibiting spirals are conducted either far away from the oscillatory threshold or under excitable resp. bistable conditions. It is tempting to explain the experiment of Vanag and Epstein [11] from the proximity of the control parameters to a Hopf bifurcation, though other more complicated possibilities cannot be ruled out at this stage.

Finally, we like to stress the most essential point in the consideration of the nature of spirals and ant spirals. The crucial quantity for both structures is the group velocity of the periodic waves outside the core region. In all models considered, the group velocity is pointing outward from the spiral core as has been shown analytically for the CGLE. The distinction between spirals and ant spirals stems only from the sign of the phase velocities and the corresponding phenomenological impression. In our opinion it is important not to confuse the distinction between spirals and ant spirals with the distinction between sinks and sources. Several authors have pointed out that the definition of a source depends only on the group velocities and not at all on the phase velocity of the periodic waves far away from it [40, 41]. Counting arguments show that sources belong typically to a zero-parameter family while sinks are members of a two parameter family [39]. In other words, sources select frequency and wavelength of the emitted periodic waves, while sinks simply appear where two wavetrains of arbitrary wavelength and period happen to meet. A classification of defects in reaction-diffusion media can be done according to the group velocities of periodic waves on both sides of the defect and the velocity of the defect itself. Apart from sources and sinks, one may also find transmission defects (one-parameter family) and contact defects in reaction-diffusion media [11]. In the present context, spirals and ant spirals are both sources, i.e. they select their frequency and wavelength and control their surroundings. If they were sinks (as sometimes claimed in the literature), the waves had to be organized from the domain boundaries, which would require another unknown “organizer” structure. Altogether, we have shown by combination of existing analytical arguments that ant spirals are structures of similar nature as spirals and should hence be expected in many systems exhibiting phase waves in particular near the onset of oscillations. Their experimental discovery can be nicely interpreted within the existing theoretical framework for oscillatory reaction-diffusion media.

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[1] R. Kapral, and K. Showalter (Eds.), Chemical waves and patterns, (Kluwer, Dordrecht, 1994).
[2] R. Imbihl and G. Ertl, Chem. Rev. 95, 697 (1995).
[3] J. Murray, Mathematical Biology, (Springer, Berlin, 1989).
[4] J. Keener and J. Sneyd, Mathematical Physiology, (Springer, New York, 1998).
[5] A. T. Winfree, Science 175, 634 (1972).
[6] G. Gerisch, Naturwissenschaften 58, 430 (1971); P. Devreotes, Science 245, 1045 (1989).
[7] S. Jakubith, H.-H. Rotermund, W. Engel, A. von Oertzen, and G. Ertl, Phys. Rev. Lett. 65, 3013 (1990); S. Nettesheim, A. von Oertzen, H.H. Rotermund, and G. Ertl, J. Chem. Phys. 98, 9977 (1993).
[8] J. M. Davidenko, A. M. Pertsov, R. Salomonsz, W. Baxter and J. Jalife, Nature 353, 349 (1991).
[9] J. Lechleiter, S. Girard, E. Peralta and D. Clapham, Science 252, 123 (1991)
[10] Th. Mair and S. C. Müller, J. Biol. Chem. 271, 627 (1996).
[11] V. K. Vanag and I. R. Epstein, Science 294, 835 (2001)
[12] T. Yamada and Y. Kuramoto, Prog. Theor. Phys., 55, 2035 (1976).
[13] J. A. Selpuchre, G. Dewel, and A. Babloyantz, Phys. Lett. A 147, 380 (1990).
[14] A. Goryachev and R. Kapral, Phys. Rev. Lett. 76, 1619 (1996).
[15] M. Ipsen M. F. Hynne and P. G. Sorensen, Int. J. Bif. and Chaos 7, 1539 (1997).
[16] S.M. Tobias and E. Knobloch, Phys. Rev. Lett. 80, 4811 (1998).
[17] A. Rabinovitch, M. Gutman, and I. Aviram, Phys. Rev. Lett. 87, 084101 (2001).
[18] M. Stich and A. Mikhailov, Z. Phys. Chem. 216, 521 (2002).
[19] S.-J. Woo, J. Lee and K. J. Lee, Phys. Rev. E 68, 016208 (2003).
[20] H. Skodt and P. G. Sorensen, Phys. Rev. E 68, 020902 (2003).
[21] I. S. Aranson and L. Kramer, Rev. Mod. Phys. 74, 99 (2002).
[22] V. K. Vanag and I. R. Epstein, Phys. Rev. Lett. 88, 088303 (2002).
[23] E. M. Nicola, W. Wolf, M. Or-Guil and M. Bär, Phys. Rev. E 65, 055101 (2002)
[24] Y. Gong and D. J. Christini, Phys. Rev. Lett. 90, 088302 (2003).
[25] L. Brusch, E. M. Nicola and M. Bär, Phys. Rev. Lett.
92, 080901 (2004).
[26] Y. Kuramoto, Chemical Oscillations, Waves, and Turbulence, (Springer-Verlag, Berlin, 1984).
[27] M. C. Cross and P. C. Hohenberg, Rev. Mod. Phys. 65, 851 (1993).
[28] G. Nicolis, Introduction to Nonlinear Sciences, (Cambridge University Press, Cambridge, 1995).
[29] M. Ipsen, L. Kramer, P. G. Sorensen, Phys. Rep. 337, 193 (2000).
[30] Y. Kuramoto and T. Tsuzuki, Prog. Theor. Phys. 55, 356 (1976).
[31] V. K. Vanag and I. R. Epstein, Phys. Rev. Lett. 87, 228301 (2001).
[32] L. Yang, M. Dolik, A. M. Zhaotinsky and I. R. Epstein J. Chem. Phys. 117, 7258 (2002).
[33] I. Prigogine, and R. Lefever, J. Chem. Phys. 48, 1695 (1968).
[34] R. FitzHugh, Biophys. 1, 445 (1961); J. S. Nagumo, S. Arimoto S. Yoshizawa, Proc. IRE 50, 2061 (1962).
[35] P. S. Hagan, SIAM J. Appl. Math. 42, 762 (1982).
[36] S. Komineas, F. Heilmann and L. Kramer, Phys. Rev. E 63, 011101 (2000).
[37] E. Bodenschatz, A. Weber and L. Kramer, in Nonlinear Wave Processes in Excitable Media, edited by A. V. Holden, M. Markus and H. G. Othmer (Plenum Press, New York, 1990); I. S. Aranson et al., Phys. Rev. A 46, 2992 (1992).
[38] J. Paullet, B. Ermentrout and W. Troy, SIAM J. Appl. Math. 54, 1386 (1994).
[39] A. T. Winfree, Chaos 1, 303 (1991).
[40] M. van Hecke, C. Storm and W. van Saarloos, Physica D 134,1 (1999).
[41] B. Sandstede and A. Scheel, SIAM J. Applied Dynamical Systems, in press (2004).