The evolution of scroll waves in an excitable chemical medium with a gradient parallel to the scroll filament is studied. Depending on the excitability and the gradient, twisted scroll waves and scroll wave attachment to the boundary of the reaction volume are observed by optical tomography, which allows the full three-dimensional reconstruction of the vortex structure in the Belousov–Zhabotinsky reaction as well as estimates on the shape and dynamics of the organizing center of the scroll (the filament). This behavior is reproduced in numerical simulations with the generic Barkley model for excitable media supplemented by a gradient along the filament. In particular, the study deals with scroll waves in a cylindrical reaction system with a free surface open to the air. Oxygen penetrating through the free surface inhibits the evolution of waves and establishes a twist of the filament. Cooling of the reaction system enhances this twist, eventually leading to a bending of the initially straight filament. Finally, the bending becomes so pronounced that the filament touches the container boundary and breaks into two pieces, each of which has its distinct rotation frequency.

I. Introduction

Excitable media play an important role in many biological systems, since they are able to support signal propagation in an intra- and extracellular context. Examples of such media are cardiac tissue,1,2 chicken retina,3 yeast extracts,4 frog eggs5 and the slime mold Dictyostelium discoideum.6 These media, if their shape is quasi two-dimensional, frequently exhibit spiral waves that rotate around a phase defect, the organizing center or core. Thin layers of the Belousov–Zhabotinsky (BZ) reaction are a well accepted chemical model system to study properties of such media. In three dimensions the spatio-temporal patterns are more complex. Examples of such patterns have been theoretically predicted by, among others, Winfree and Strogatz,7 the simplest of which are scroll waves and rings. For both structures either spatially fully resolved single snapshots8,9 or time resolved projections10,11 have been published so far.

Scroll waves can be viewed as a stack of spirals, all having the same frequency. For a simple scroll wave all the spirals possess the same phase, otherwise it is called twisted. The rectilinear line connecting the cores of all these spirals is named the filament. In a cylindrical reaction system this line may reach from one base to the other. Scroll waves have been studied theoretically out of two main motivations: The first aim is to provide additional insight into particular systems like the slug of the slime mold Dictyostelium discoideum12 or electrical activity in cardiac tissue.13 The second direction is the study of vortex filaments in generic oscillatory and excitable media. Popular models are the complex Ginzburg–Landau equation for the oscillatory case14 and the FitzHugh–Nagumo model in the version suggested by Barkley for the excitable case.15 Furthermore, issues of geometry16 and heterogeneities17,18 have been explored for excitable media.

Recently, progress in the analytical understanding of the selection of twisted scroll waves15 and a detailed stability analysis19 of scroll waves in isotropic and homogeneous media was obtained. Margerit and Barkley derived rigorously that an increase in twist leads to an increase in the selected frequency of the scroll wave.13,20 Hakim and Henry showed by a numerical stability analysis that filaments can have a negative line tension,21 which leads to spontaneous bending of the filament and, in a finite box, scroll wave attachment to the boundaries. Scroll waves in anisotropic and heterogeneous media have been predominantly studied by numerical simulations. Mikhailov et al. used also the kinematic theory in an early study of a step heterogeneity to explain that the scroll wave gets twisted in the part with weaker excitability.22

Simulation studies of scroll waves in heterogeneous media have shown attachment for large gradients and twist for small gradients.23 It is instructive to discuss these observations in terms of synchronization. Regions in the reactive medium with weaker excitability, where spirals in a corresponding two-dimensional system rotate with slower frequency, may compensate this, in the case of a small gradient, by twisting. Too much twist, however, leads to destabilization and bending of the straight filament.14,21

If the filament touches the boundary, it splits into two parts, as observed in BZ experiments by Mironov et al.24 The frequencies of the two fragments of the original filament are then no longer synchronized and often the one with the higher frequency suppresses the other filament. As a result one single filament persists that connects the end of higher excitability with the lateral surface of the reaction cuvette in experiments, or with the lateral surface of the cylindrical simulation box in
numerical calculations. The remaining fragment emits wave sheets towards the opposite end of the cylinder. These are stretched into almost planar waves while they move into the direction of weaker excitability, where some of them, however, are wiped out. The latter observation is reminiscent of behavior seen in a two-dimensional medium with a step gradient, where not all waves emanated from a core in the region of higher excitability succeed to propagate into the other half of the medium. The present paper reports on spatially and temporally resolved observations of scroll waves in a three-dimensional BZ reaction system that are exposed to a gradient of excitability. The dynamics of a filament reaching from one base of a cylinder to the other and the three-dimensional shape of the waves are reconstructed with the help of optical tomography. The method allows to measure local quantities like oscillation frequency and wave speed. In particular, in a vertically aligned cylindrical cuvette, oxygen is penetrating into the reaction system across the open top surface, thus inducing a gradient of excitability. Scroll wave attachment results from collision of a part of the filament with the lateral surface of the container. Situations where two scroll fragments coexist and where one scroll fragment emits quasi-planar wave sheets are visualized. The scroll wave attachment is found to desynchronize local oscillations in different parts (bottom and top) of the cuvette. We corroborate the experimental findings by three-dimensional numerical simulations with the Barkley model supplemented by a vertical gradient of excitability. Different gradients are investigated. Scroll wave attachment is more likely to occur for sudden step-like gradients than for smooth linear gradients of the parameters. The desynchronization is found to occur for situations when the point at which the filament touches the lateral surface either rotates around the boundary or is pinned to a heterogeneity near the boundary. The simulations reproduce most aspects of the experiments and show various regimes of wave penetration from the filament fragment near the bottom to the upper half of the medium including 1 : 1-, 1 : 2-frequency entrainment, and quasiperiodic response.

II. Materials and methods

A. Chemical preparation

Experiments were carried out with the ferroin-catalyzed BZ reaction. The reactants and their concentrations are listed in Table 1. Due to the fact that the three-dimensional geometry of the investigated system is an essential feature of the experiments, all reactants have to be added to the system from the very beginning. In order to avoid any hydrodynamic effects perturbing the wave fronts the reaction was embedded in an agar gel matrix. Furthermore, to reduce the production of CO₂ bubbles, the surfactant sodium dodecyl sulfate (SDS) was added at a concentration below the critical micelle concentration (cmc ≈ 4.0 mM). Agar (purified agar, Sigma) was 0.075% by weight, which is slightly above the limit required for gelation (with 0.070% w/w the reaction system remains liquid even at 20°C).

For preparation of 10 ml of the solution the agar was mixed with water in a beaker and heated until it was clear and without streaks (at ≈90°C). SDS was added, and after the solution had cooled down to 50°C, the water lost by evaporation was compensated. The components of the BZ reaction were added at 36°C, just before the solution was poured into the cylindrical cuvette. In the further course of the experiment the system settled on the temperature controlled by the cryostat.

Table 1 Table of reactants

<table>
<thead>
<tr>
<th>Solvent</th>
<th>Concentration</th>
</tr>
</thead>
<tbody>
<tr>
<td>Malonic acid (Riedel-de Haën)</td>
<td>50 mM</td>
</tr>
<tr>
<td>Sodium bromate (Fluka)</td>
<td>50 mM</td>
</tr>
<tr>
<td>Sulfuric Acid (Riedel-de Haën)</td>
<td>200 mM</td>
</tr>
<tr>
<td>Ferroin (Fluka)</td>
<td>0.5 mM</td>
</tr>
<tr>
<td>SDS (Fluka)</td>
<td>0.05 mM</td>
</tr>
</tbody>
</table>

B. Initiation of waves

For initiation of the wave structure the partition method was used. 8.4 ml of the reaction solution were filled into the reaction cuvette (Quartz glass Suprasil, Heraeus, outer diameter = 20 mm, inner diameter = 18 mm, height = 70 mm). The reaction volume was divided into two compartments by a glass plate of 0.15 mm thickness (Fig. 1), the edges of which are covered with a teflon film. This way waves were prevented from slipping through a gap between the glass plate and the reaction cuvette. A cylindrical wave was initiated with a silver wire (diameter = 0.2 mm) close to the contact line between the glass plate and the cuvette. When this wave front hits the glass plate, a segment of the cylindrical wave persists, building two contact lines with the glass plate (Fig. 1). These lines then move away from the ignition site, one towards the center of the cuvette, the other towards the contact line between glass plate and reaction cuvette. After the latter crosses this contact line, the glass plate is carefully removed so that the gel can close the gap, which was induced by the removal of glass plate. The first, inwardly moving line is left as open wave edge in the bulk of the reaction volume and will curl in to form a scroll wave. The filament of the scroll wave is located close to the position of the first contact line at the moment when the glass plate was removed.

C. Tomography

For observation, the reaction cuvette subsequently is placed into the tomographic setup (Fig. 2). For illumination a blue LED (λ = 470 nm, bandwidth = 50 nm) is utilized, so that further optical filters are not necessary. One tomographic sample (sinogram) consists of 100 projections through the reaction cuvette taken from different angles. Between two adjacent projections, a stepper motor rotates the probe by an angle of 1.8°.

Thus, one sample contains projections from different angles which are equally distributed over the semi circle. Recording is done with a CCD-camera (Hamamatsu, 25 frames s⁻¹). Its signal is digitized by a framegrabber-card (Data Translation, dt3155). It takes 4.0 s to record one sample, and therefore, with the given spatial resolution of 0.1 mm/pixel and the measured
velocity of less than 0.25 mm s⁻¹, the wave structure can be considered frozen-in during the acquisition of the projections of the sample. However, noise reduction by temporal averaging was not possible.

A necessary condition for reconstructing the spatial distribution of the absorption inside the cuvette is a parallel light beam. To obtain such a pathway of the light a lens is used and the cylindrical reaction cuvette is immersed into a square-shaped cuvette for index matching. The water in the square-shaped cuvette is connected to a cryostat and serves simultaneously as thermic bath for the reaction.

Spectrophotometric considerations show that the recorded samples of the reaction volume are slice-wise the Radon transform of the ferroin/ferriin concentration. Therefore, the whole information about the spatial concentration distribution is contained within the samples, however, local quantities are not directly accessible. Mathematically the spatially distributed concentration of ferroin/ferriin inside the cylindrical cuvette, \( c(x, y, z) \) with \( x^2 + y^2 \leq R^2 \) and \( 0 \leq z \leq H \) (\( R \) = radius of the cuvette, \( H \) = height of the reaction solution), is slice-wise mapped onto a function \( R(\phi, \psi, z) = \int_{c(x, y, z)} \cos \phi \sin \psi \, dx \, dy \, dz \) with \( \cos \phi \sin \psi \) the distance \( r \) from the center of the cuvette. For the derivation of local quantities one has to apply techniques as discussed in refs. 30,31 to the sample data to invert the Radon transform.

It was tested by recording and reconstructing different suitable objects of known size that the resolution of this observation method is satisfactory. Especially, the distortion of space due to diffraction at the cylindrical cuvette was negligibly small. Furthermore, the gel was stiff enough to avoid blurring of edges due to vibrations induced by the stepper motor. This was tested by immersing thin wires into the gel. The size of the wires and their distances from each other could be correctly reconstructed.

### D. Measurement of local quantities

The raw data recorded during the experiment consists of samples of 100 projections of the reaction volume. One example of a projection is given in Fig. 3, center image. Taking into account at least two directions of projections they allow a qualitative estimate of phenomena like e.g. the bending of the filament (when using only one direction the bending could occur in that direction, and therefore could be lost in the recorded data).

For the observation of localized quantities like time series of concentrations at a given point one has to apply the reconstruction technique to all the projections of a sample. The result of tomographic recording is a stack of slice-wise reconstructed concentration data, where the localization of the concentration data is given by the spatial resolution of the reconstruction algorithm. Fig. 3, right, shows four slices of such a stack. Time series of concentration data were obtained by selecting given points in the volume and collecting the gray levels at these locations in the course of time. (In the following we call this series gray level time series.) Frequencies at different locations were measured by applying a Laplace of Gaussian (LoG) filter to these data (see 33). Waves passing through such locations are detected by the zeros of the filtered time series.

To obtain the horizontal wave velocity we extract slices of the reaction volume that are perpendicular to the axis of the cylindrical cuvette. (The spirals in these slices (see Fig. 3, right) are given by the intersection of the slices with the scroll wave, the core is given by the intersection of the slices with the filament.) With horizontal velocity we then denote the velocity of the “intersection wave” in this two-dimensional slice. For measuring it at a certain height, a line is fixed in the slice at that height such that it is both perpendicular to the propagation direction of the intersection wave and collinear to the movement of the intersection core. The gray levels along this line are LoG-filtered. The zeros of this filtered data give the location of the intersection wave along this line. Tracing these locations in the course of time allows us to calculate the velocity of the intersection wave trains. One should mention that determining the locations of the wave trains by applying a LoG-filter is suitable under the condition that the wave trains have a stationary profile.

Isoconcentration surfaces are calculated by applying the marching cube algorithm to the reconstructed data stack. A perspective representation of such an isoconcentration level is shown in Fig. 3, left. Movies of such presentations give an excellent impression of the scenario in the interior of the reaction volume. They allow an easy qualitative understanding of the behavior of the wave structure. The decomposition of an isoconcentration level into several connected components (see Fig. 6) is a pronounced change in the wave behavior and can be easily detected using this presentation.

### III. Experimental results

Periods and phases of gray level time series and horizontal velocities of wave trains at different heights of the reaction volume are used to determine characteristic properties like bending and twist of the organizing filament. Due to a variation of the temperature of the reaction system the observed wave structure develops from a twisted scroll wave towards a wave structure with a bent filament emitting quasi-planar wave sheets.

#### A. Estimates on filament dynamics: initial stage

At the beginning of the experiment the temperature is kept at 22.0°C. After a short equilibration time a fixed twist is
The initial value is 22 °C, the plot shows the evolution of the temperature in the thermic bath. The jump of the amplitude at 4500 s stems from a readjustment of the illumination.

Established as clearly demonstrated by the time series of projections (Fig. 3, center). The twist is not equally pronounced at all height levels but is located near the top base of the cylinder containing the reaction system. At this stage periods of the gray level time series remain the same at all heights of the volume (Fig. 4, $t \leq 6000$ s). This is also reflected in Fig. 5 which shows the temporal evolution of the periods of the time series. There is, however, a phase shift of about $\pi$ between the top and bottom of the reaction system (Fig. 4). The shape of isocentrification surfaces of the wave structure remains constant for several rotations as can be seen in time series of perspective representations of the wave structure. (A snapshot of such a time series is shown, for instance, in the left of Fig. 3.) Furthermore, the horizontal wave velocity is nearly the same throughout the cuvette at this stage. Since the fronts in the projections are nearly vertical (Fig. 3, center), i.e., perpendicular to the slices, this velocity of intersection waves can be taken as the full three-dimensional velocity of the fronts of the scroll wave. The constancy of the horizontal velocity indicates that the twist is small and any bending of the filament is negligible if present at all. At this time, therefore, the filament is a straight line. These observations show that the twisted scroll wave in the chosen reaction system is a stable structure at 22.0 °C.

### B. Estimates on filament dynamics: intermediate stage

The excitability of the reaction system is then decreased by cooling the solution from 22.0 °C to 11.0 °C during a time period of 50 minutes. This process induces a temperature gradient of at most 0.5 °C cm$^{-1}$ in the radial direction between the lateral surface and the center of the cuvette but has no vertical component. We argue that in our cylindrical cuvette of 18.0 mm inner diameter this gradient has no significant influence on the behavior of the filament: In ref. 18 a scroll wave was exposed to a gradient of excitability parallel to the filament. It has been found that in this situation the filament tends to move on a pathway with a component perpendicular to the gradient. In our situation the radial gradient could be expected to force the filament on a pathway which possesses a circular component. No indication of such motion is observed, therefore effects due to such a radial gradient can be assumed negligibly small.

As a further consequence of the cooling process the rotation of the scroll wave slows down yielding roughly four times longer periods (Fig. 5). The previously observed phase locking of the gray level time series disappears. The phase of the gray level time series measured in the inhibited top region lags increasingly behind that of the bottom. After about 90 minutes the phase shift between the top and bottom end has grown by $\pi$ (Fig. 4). Projections through the reaction volume and stacked slices show an increasing twist.

It is important to note that the increasing phase shift of the gray level time series is not only due to an increasing twist: the initially straight filament also starts to bend in the upper part of the cuvette, as can be seen from the perspective representation of an isocentrification level (Fig. 6) and stacked slices (Fig. 7). Thus, the core of the spiral in the upper slices, given by the intersection of the filament with these slices, starts to move. Therefore the phase shift is also due to a Doppler effect.

This evolution demonstrates that decreasing the temperature induces a qualitative change of the influence of the open surface. Oxygen is known to play an inhibiting role in the ferroin-catalyzed BZ reaction and is the main candidate to explain the observed system behavior. It has been reported that the inhibiting effect of oxygen may reach up to 1.3 mm below the surface of a ferroin-catalyzed BZ reaction and that it becomes the more pronounced the lower the excitability of the reaction system is. Given that the reactivity of oxygen towards the BZ reaction depends on the temperature, we conjecture that variations in temperature effectively change the depth of solution penetrated by oxygen: the higher the temperature the higher the reactivity of oxygen towards the BZ reaction and consequently the higher the consumption of O$_2$ close to the interface. In other words, at higher temperatures O$_2$ is more effectively sequestered from the reaction mixture, leading to a lower or shallower “penetration depth” of oxygen. Thus, this may explain why the bend appears at a depth of 3.5 mm–8.5 mm when the system is cooled to 11.0 °C.

### C. Estimates on filament dynamics: final stage

When the bending of the filament begins, the emitted wave fronts are no longer vertically aligned. The velocity has a vertical component which changes the relationship between the full three-dimensional velocity and the horizontal velocity. Vice versa, measuring the horizontal velocity near the organizing center allows an estimate of the bend of the filament. The horizontal velocity $v$ can be computed according

---

**Fig. 4** Time series of gray levels measured at fixed horizontal positions in slices at top and bottom of the reaction cuvette (dashed = 3.5 mm, solid = 28.5 mm from the top). The dotted line at the bottom of the plot shows the evolution of the temperature in the thermic bath. The initial value is 22 °C, between 4800 s and 7800 s the temperature is lowered to 11 °C. The period in time where phase locking is observed is shaded. The jump of the amplitude at 4500 s stems from a readjustment of the illumination.

**Fig. 5** Periods of selected oscillations of the gray level time series measured at a fixed horizontal position at different heights. Corresponding oscillations which stem from the same revolution of the scroll wave are connected by lines. Numbers indicate the number of the revolutions. Heights of the positions are $\bigsquare \approx 3.5$ mm, $\bigcirc \approx 8.5$ mm, $\bigtriangleup \approx 13.5$ mm, $\blacktriangle \approx 18.5$ mm, $\triangleleft \approx 23.5$ mm, and $\blacktriangledown \approx 28.5$ mm from the top.
to \( v = v_0 / \sin(z) \), where \( v_0 \) is the velocity of the wave fronts and \( z \) is the angle enclosed by the propagation direction of the waves and the vertical. Close to the filament the propagation direction is perpendicular to the filament. Geometrical considerations then show that it is possible to draw conclusions from the horizontal velocity to the tilt of the filament against the vertical. The horizontal velocity starts to grow in the upper slices after the cooling process (Fig. 8). Thus there arises a bending of the filament which is most pronounced near the top. With the horizontal velocity increasing by a factor of about three in the topmost slice, the maximum tilt of the filament is at least \( 70^\circ \).

When the filament touches the boundary two desynchronized fragments remain, each of which has its own frequency and dominates a certain domain of the reaction system. In a vertical slice intersecting with both filaments a pair of counter rotating spirals is observed. Due to this splitting of the organizing line there is a sudden jump of the period of the gray level time series of about 10\% in the topmost plane (Fig. 5). It stems from the second fragment that rotates significantly slower and appears only temporarily (for at least two revolutions). Subsequently the measuring point in the topmost slice belongs to the part of the system that is dominated by the faster fragment of the filament in the bottom region of the cuvette. Time series of perspective representations of isoconcentration levels finally show the emission of planar wave sheets from bottom to top (Fig. 6). In horizontal slices through the cuvette then nearly homogeneous oscillations occur instead of spirals. The oscillations stem from the quasi-planar wave sheets passing through that slice. The disappearance of the slow filament is reminiscent of the behavior of two counter rotating spirals with different frequencies seen in a two-dimensional medium, where the slower rotating spiral is driven away by the faster one.\(^{37}\)

IV. Simulations

A. Model

A standard model for the Belousov–Zhabotinsky reaction is the two variable Oregonator.\(^{38}\) Qualitatively, it is very similar to generic models for excitable media, such as the FitzHugh–Nagumo\(^{17}\) or the Barkley equations.\(^{39}\) Although the latter is not a model derived explicitly for the BZ reaction, we use it, because it is computationally the most efficient one and allows systematic simulations in three spatial dimensions. Thus, in our model calculations there are no direct relations between the variables \( u \) and \( v \) and the concentrations of individual chemical species involved in the BZ system. In a loose analogy, the variable \( u \) here corresponds to the autocatalytic species in the BZ reaction, while \( v \) plays the role of the slow inhibitor. The numerical methods used have been described by Dowle \textit{et al.}\(^{40}\) and the implementation required only a minor modification of the EZSCROLL software.\(^{40}\) The explicit model equations are:

\[
\frac{\partial u}{\partial t} = f(u, v) + \nabla^2 u \tag{4.1}
\]

Fig. 6: Time series of perspective representation of isoconcentration levels of the full three-dimensional reconstruction. Times are (left to right) 110 min \( \pm \) 18 revolutions, 176 min \( \pm \) 29 revolutions, and 256 min \( \pm \) 34 revolutions after ignition of the scroll wave.

Fig. 7: Time series of horizontal slices through the reaction cuvette. Locations of the slices are the same as in Fig. 3, times are the same as in Fig. 6. The filament is shown as a dark line.

Fig. 8: Horizontal velocity of wave fronts in different slices. Heights of the slices are the same as in Fig. 5. The cooling process (7800 s) decreases the horizontal velocity. After the cooling process it increases significantly in the upper slices.
where \( u \) and \( v \) are known as the excitation and recovery variables, respectively; \( f(u, v) \) and \( g(u, v) \) express the local reaction kinetics; and \( D \) is the ratio of the diffusion coefficients.

The local kinetics is given by:

\[
f(u, v) = \frac{1}{e} u(1 - u)(u - u_h(v))
\]

and

\[
g(u, v) = u - v,
\]

where \( e \) is a small parameter and

\[
u_h = (v + b)/a
\]

is the excitability threshold. The model has three free parameters: The positive quantities \( a < 1, b < 1, \) and \( e < 1 \) are chosen to realize an excitable medium with the stable uniform rest state \((u_0, v_0) = (0, 0)\). The variable \( u \) represents a fast activator species, while \( v \) is the slow inhibitor that ensures a feedback and the return to the rest state. Here, we incorporate the effect of a heterogeneity into the model by replacing the parameter \( b \) by a function \( h(z) \) and study its effect on a scroll wave whose filament is initially oriented along the \( x \)-axis. The simulations have been performed in a rectangular box with typical extensions \( L \times L_x \times L_z = 40 \times 40 \times 60 \) and zero-flux boundary conditions. To mimic the cylindrical geometry of the experiment, we increase the excitation threshold \( v \), which in turn slows down or even suppresses the propagation takes only in a cylinder embedded in the rectangular box.

B. Results

For the simulations, we fixed all parameters except \( b \). Effectively, increasing \( b \) leads to a higher excitation threshold, which in turn slows down or even suppresses the propagation of excitation waves. The vertical heterogeneity will be taken into account by choosing a \( z \)-dependent \( b \)-value. As a first step it is instructive to study the change in the spiral wave properties in two dimensions with the parameter \( b \). The dependency of the spiral period \( T \) on \( b \) is displayed in Fig. 9 for three different choices of the diffusion constant \( D \). In all cases, we see the expected increase of the period with \( b \) and finally a divergence at a value \( b = b_{crit} \) beyond which spirals do not form anymore. Following ref. 41, we will use a value of \( D = 0.6 \) estimated from the molecular weights of the reactants in the BZ reaction. The results, however, do not sensitively depend on this choice; they are just a consequence of the dependence of the period on \( b \) values. A vertical gradient in \( b \) leads to a vertical gradient in the rotation period of the in-plane spiral that can be compensated by a twist of the scroll wave. For the Barkley model in homogeneous media and at sufficiently small \( e \), the selected frequency of a twisted scroll wave has been derived analytically for the case \( D = 0 \), see 42, and reads

\[
\omega_0 = 0.692 \mu (a, b) e^{-1/3} - 0.926a^{-1} - a_1 (a, b) v^2 + O(e^{1/3}),
\]

where \( \omega_0 \) measures the twist of the wave. The coefficient \( a_1 \) depends on the original parameters \( a \) and \( b \) and is always negative. This result implies that twist increases the rotation frequency. A similar behavior is seen for scroll waves whose filament is aligned parallel to the \( b(z) \)-gradient in a medium. Even if the initial scroll wave is untwisted, the different rotation frequencies in the respective \( x \)-\( y \)-planes will immediately introduce a difference in the phase of the rotation along the \( z \)-axis and hence a nonuniform twist in the scroll wave. This twist, however, leads to an increase of the rotation frequency and may even compensate the shift in rotation frequency along the \( z \)-axis introduced by the gradient. In our simulations, we find that the scroll wave twists in a way that synchronizes the oscillations in the medium and leads to the same rotation frequency in each plane - this is in agreement with the experimentally found time series near the top and bottom of the reaction vessel in Fig. 4. The twist leads also to a bending of the initially straight filament of the scroll wave. If the bending amplitude becomes too large the filament may get attached to the system boundary and the scroll wave then "breaks" at this point and emits almost planar waves into the medium in accordance with the experimental data presented in the first part of this paper and the earlier observation of Mironov et al.24

![Fig. 9](image)

**Fig. 9** Period dependence on parameter \( b \): for \( D \neq 0 \) the spiral period is smaller than in the no diffusion limit. Model parameters: \( a = 0.8 \) and \( 1/e = 150 \). Numerical parameters: \( L = 50, N = 161, t_e = 0.8, \) and \( \delta = 0.001 \).

We studied two kinds of \( b \) gradients in the \( z \) direction: a linear and a step gradient, defined by

\[
b(z) = b_{min} + (b_{max} - b_{min}) \frac{z}{L_z}
\]

for the linear gradient and

\[
b(z) = b_{min} \text{ if } z < z_i \text{ and } b(z) = b_{max} \text{ if } z \geq z_i
\]

for the step gradient. For each type of gradient, we varied the value of \( b_{max} \), see Table 2.

Snapshots of the wave dynamics for \( u = 0.6 \) are shown in Fig. 10. While there is scroll attachment for large linear \( b \) gradients, there is no resulting complex wave activity in the upper part of the medium. Instead, e.g. for the largest linear \( b \) gradient (first row, right picture), the \( b \) value surpass the limit of wave propagation and wave activity rapidly decays above that point leaving a quiescent top layer. For the smallest step \( b \) gradient (second row, left picture), we observe a twisted scroll in the simulation. Filament interaction with the boundaries is

<table>
<thead>
<tr>
<th>Gradient Type</th>
<th>( b_{min} )</th>
<th>( b_{max} )</th>
<th>( b_{max} )</th>
<th>( b_{max} )</th>
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<td>Linear</td>
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<tr>
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</tbody>
</table>

responsible for scroll fragmentation and planar like wave emission, that happens for the higher step $b$ gradients. This has been confirmed by increasing the cuvette diameter from $L = 40$ to $L = 60$, which changes the critical $b_{\text{max}}$ value at which the filament breaks. We reconstructed the approximate dynamics of the filament by analyzing the intersection of the level curves $u = 1/2$ and $v = a/2 - b$. This intersection is a reasonable definition of the spiral tip. Filament snapshots for these scrolls are shown in Fig. 11. For the step $b$ gradient, fragments of the filaments can be seen in the middle and left frame in the bottom row of Fig. 11 after the filament's attachment to the boundary.

The temporal evolution of the scroll wave can be seen in the space-time portraits for the plane $y = L/2$. Fig. 12 shows the evolution of the scroll for the smallest step $b$ gradient in Table 2. There is no scroll attachment and the oscillations in the top and bottom of the scroll wave are synchronized, i.e. they have a constant phase relation. The tilted waves visible in the $x = 40$ plane indicate that the scroll is twisted, as required for synchronization.

For scroll dynamics at larger step $b$ gradients, see Fig. 12 center and bottom, the space-time portraits indicate the scroll attachment, compare the broken wave fronts in the $x = 40$ plane. In both cases, the scrolls are twisted, but they are not synchronized anymore. Finally, we studied the case where the filament gets pinned by a lateral defect with low excitability (large $b$ value) after attachment to the boundary. In the medium without heterogeneity the filament is seen to move around the boundary as in similar observations in earlier model calculations. The experimental data from the BZ experiment, however, show that the filament is pinned to one particular place to the boundary. Comparison of the simulation shows however that this doesn’t affect the main characteristics of the transition from a twisted scroll wave to a scroll wave attached to the boundary. It has been noted earlier that this transition is accompanied by irregular behavior. Our measurements above clearly show that the synchronization between the lower half of the medium governed by the surviving part of the initial scroll and the upper part characterized by sheets of planar waves is lost. Fig. 12 and Fig. 13 show that this is due to the fact that not every wave emitted from the scroll succeeds to enter the upper half of the medium. While part of the time evolution suggest a 1:2-transmission of the waves into the upper half analogous to the two-dimensional case studied in, in general there is no clear relation between waves in the lower and

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**Fig. 10** Typical scrolls for surfaces of $u = 0.6$ for the linear gradient (top line) and for the step gradients (bottom line) at time $t = 225$, after 3000 time steps. There is scroll breakup only for high enough step gradients. The values of $b_{\text{min}}$ and $b_{\text{max}}$ are given in Table 2. Model parameters: $a = 0.8, 1/e = 50, L = 60$ and $D = 0.6$. Numerical parameters: $N_x = 81, N_y = 81, N_z = 121, t_s = 0.8$ and $\delta = 0.001$. 

upper part which leads to the observed desynchronization. Altogether, the simulations do reproduce the behavior found in the experiment described in the previous section and are a valuable tool to understand the details of the transition from twisted scroll waves via scroll wave attachment to complex behavior and desynchronization of the medium.

V. Discussion and conclusions

In this work the behavior of scroll waves under the influence of a localized, nearly step-like gradient was investigated experimentally and in simulations. Previous studies have shown this type of gradient to be an important system property, e.g., in the formation of the slug of the slime mold Dictyostelium discoideum.12 Other than in the experimental design given in ref. 24, temperature was used as an indirect control variable. A comparison of projections through the reaction volume clearly reveals the difference in the location of twist along the vertical axis. In the present study it is localized (cf. Fig. 3), which is due to an inhibiting effect near the surface. Decreasing the temperature strengthens the localization and the steepness of this inhibiting effect and therefore leads to an experimental approximation of a step-like gradient of excitability. This change in shape of the gradient will force a scroll wave to behave in the observed manner.

Simulation results based on a generic model for excitable media reflect in good agreement the experimentally observed system behavior. Introducing a step-like increase of the $b$-value of this model (i.e. increasing the excitation threshold) to decrease the excitability in a certain part of the simulation box reproduces the system response to a decrease of temperature in the experimental ductus. The simulations distinguish clearly between the behavior induced by a linear gradient and that caused by a step-like gradient. By taking into account, for instance, time series of the activator variable measured in the bottom and top region of the simulation box one sees that for both cases the influence of the remaining filament on the weakly excitable region is distinct. Topologically the final stages of the simulated wave structures are also different in the way that with the step-like gradient several fragments of the filament may form that act as frequency sources just as we observe in the experiments. This correspondence, in fact, gives a strong hint that with the chosen experimental design a more step-like gradient of excitability is realized.

Recently, bending has been explained to occur due to negative tension of the filament. 43 Simulations with the Barkley model again exhibit a region of excitability, where filaments of scroll waves in a homogeneous system tend to stretch themselves, form loops and complex tangles, eventually leading to “Winfree turbulence”. 43 This negative tension as a reason for the observed filament instability is not supported by further experiments performed in our laboratory. For the given concentration of reactants scrolls with initially curved filaments tend to linearize their organizing center. This has been observed on scrolls with a U-shaped filament connecting two points of the lateral surface of the cylinder, and therefore not reaching the top surface (results not shown). During several revolutions they develop towards a scroll wave with a rectilinear filament, where during the observation time the temperature evolution was controlled in the same way as in the experiments reported here. System aging as a reason for the observed behavior was excluded by varying the duration of the initial stage, before the excitability was manipulated by decreasing the temperature. It turned out that the bending of the filament could be observed independently of the duration of the initial stage. Furthermore, simulations showed that the existence of a small localized inhomogeneity (possibly due to the preparation procedure) does not change the characteristic system behavior during the transformation of a twisted scroll wave into a bent structure sending out quasi-planar wave sheets (Fig. 13).

In further studies it would be challenging to investigate the scroll filament dynamics. Reconstruction of the filament by an overlay technique could be a first step in that direction.
Deeper insight into the nature of the filament instability can be expected by the evaluation of geometrical properties of the instantaneous filament as defined in ref. 11 which corresponds to the tip of a spiral in two dimensions. For the reconstruction of this instantaneous filament, optical tomography, as used in this publication, is a highly suitable tool.

Fig. 12 Time evolution of the scroll at plane $y = L/2$ for the smallest (top), for the intermediary (center), and for the highest (bottom) $b$ step gradient: $(b_{\text{min}}, b_{\text{max}}) = (0.02, 0.065)$, $(0.02, 0.094)$, and $(0.02, 0.107)$ respectively.

Fig. 13 Time evolution of the scroll at plane $y = L/2$ for the intermediary (top) and for the highest (bottom) $b$ step gradient and a lateral defect with $b = 0.02$.

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References

32 Coloured versions of Figs. 3, 6, 7, and 10 are available at http://www.uni-magdeburg.de/abp/jpeg_colour/scroll.html, 2003.