Forced parallel drift of spiral waves in the Belousov-Zhabotinsky reaction

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The dynamic behavior of spiral-shaped excitation patterns in the Belousov-Zhabotinsky reaction was studied under the influence of externally applied weak directed current up to the order of 20 mA. A parallel drift of the centers of a symmetric pair of counterrotating spirals was observed. Furthermore, the relaxation of the spiral core locations after switching off the current was investigated. The experimental results were reproduced in numerical simulations with a reaction-diffusion model. [S1063-651X(97)06803-7]

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I. INTRODUCTION

Spatiotemporal patterns formed in systems far from equilibrium have been intensively investigated in recent years. One of the most interesting and thoroughly studied systems remains the Belousov-Zhabotinsky (BZ) reaction [1], which is the oxidation of malonic acid by bromate in the presence of catalysts such as ferroin, ruthenium, or cerium. Extended excitable systems, as realized in thin layers of the BZ-reaction solution, exhibit patterns such as rotating spirals or expanding circular trigger waves [2,3]. In particular, the motion of the tip of a spiral and the possibility to control this motion by means of weak external forces are a major focus of recent scientific interest [4–6]. For BZ media showing a sufficiently high excitability, the trace of the spiral tip is a circle, the location of which remains stationary in time [7]. The path of the tip can be influenced by changing the excitability locally or globally or by introducing a spatial gradient. The common methods to do this are using light to illuminate a photosensitive variant of the BZ reaction [8,9], which leads to a change in the excitability of the system, or applying an electric field [10] which introduces a drift of ionic key species of the reaction. As known from previous studies, an externally applied electric field causes a drift of the center of a spiral not only in the direction parallel but also in the direction perpendicular to the field [11,12]. Let us look at a thin sample layer from above: For a field orientation in which the cathode is on the left and the anode is on the right, a clockwise-rotating spiral in the upper portion and a counterclockwise-rotating spiral in the lower portion of the system will be pulled together. Thus this effect can be used to force an interaction between the spirals by reducing their mutual distance. In previous studies [13,14] the effect of a relatively high electric field (1.68–8.6 V/cm) applied to symmetric and nonsymmetric arrangements of pairs of spirals with respect to the direction of the electric field was investigated. An annihilation of spiral pairs could be observed when the symmetry axis of the spiral pair was nearly parallel to the electric field. Spiral pairs with a different initial orientation and a phase shift in their respective rotation interacted for a short time and continued then to drift in their previous direction [5].

In this work we used a weaker electric field ($E \leqslant 2$ V/cm) to study the repulsing force between two spirals in an arrangement, where the spiral pair is symmetric with respect to the vector of the electric field. Because of the different arrangement of the electrodes and different thickness of the reaction medium, this leads to a current, the strength of which is comparable to the current used in the abovementioned studies. In the following we will refer to the current as the control parameter, characterizing the size of the perturbation.

For the visualization of the behavior of the spiral pairs a different method was applied, which allows one to follow the trajectory of the spiral tip automatically. This is a significant improvement of earlier studies, which focused mainly on the observation of subsequent positions of the spiral core [12,14].

II. MATERIALS AND METHODS

Experiments were performed under batch conditions in a reactor similar to that described by Steinbock, Schütze, and Müller [12] (Fig. 1). The BZ-reaction system was embedded in a 0.4% agar gel solution, which after solidification prevents hydrodynamic disturbances. The initial concentrations of the components were 0.05M $NaBrO_3$, 0.2M H_2SO_4, 0.05M $CH_2(COOH)_2$, and $6.25\times10^{-4}M$ ferroin-sulphate $[Fe(phen)_3SO_4]$ as catalyst and indicator. A volume of 4.5 ml was placed in a flat rectangular dish (30×62.5 mm^2) yielding a layer thickness of 2.4 mm. Under these conditions patterns persist for more than 2 h, with a gradual change in the period of spiral rotation from 60 ± 5 s in the beginning to

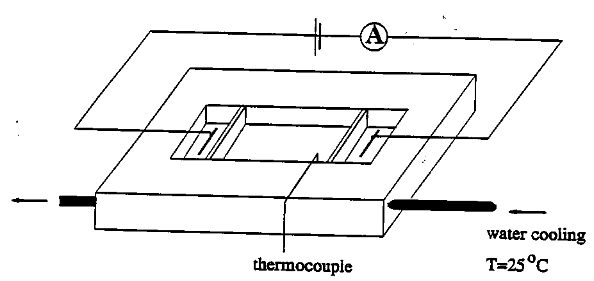

FIG. 1. Schematic representation of the reactor used in the experiments. Two reservoirs filled with saturated K_2SO_4 solution are separated from the gel in the center by porous glass. In either of the two reservoirs two platin wires serving as electrodes are immersed.

75±5 s after 60 min. In order to prevent heating of the medium due to the electric current the temperature of the dish was controlled with a cooling box at 23(±0.5) °C. About 5 min after starting the reaction by adding the catalyst, a circular wave was triggered by touching the surface of the layer with a thin silver wire. A pair of spirals evolved when part of the wave front was stopped and disrupted by a vertically inserted, removable thin glass plate (0.1 mm thick). No visible damage of the gel matrix was observed because of the low agar concentration, which results in a very soft gel. In order to produce a symmetric pair of counterrotating spirals having the desired orientation with respect to the electric field, it was necessary to ensure that the wave front and the glass plate were exactly parallel and perpendicular to the electric-field vector. The electric field was applied to two electrodes as shown in Fig. 1. A dc power source, used in constant current mode, supplied a current I in the range 0 mA$\leqslant I \leqslant$30 mA. Images were recorded with a charge coupled device camera (Hamamatsu C3077) on a video recorder and later digitized on a personal computer with an image acquisition card (Data Translation 3851 B8) and stored on a hard disk for further computational treatment.

To analyze the digitized images the quality of the images was enhanced by applying methods of digital image treatment. First, the background of the images was removed by the following method: We calculated a background image by adding up the values of the corresponding pixels in all images of the digitized movie (typically about 500) and dividing the result by the number of images. The background image was then subtracted from each single picture of the digitized movie. This way the defects that are not time dependent, such as dust particles or bubbles, can be almost completely removed. One should notice that this method works successfully because the waves are crossing each pixel in each image for about the same number of times, so that the averaging procedure removes the spirals from the background image. It is just necessary to compensate the higher overall gray level of the background image by shifting the gray levels of the images after the substraction into the positive range of gray values ($\leqslant$255).

Subsequently, the trace of the spiral tip was followed automatically with a program written in IDL [15]. For this purpose we defined the spiral tip as the intersection of two isocontour lines at the same gray level obtained from two subsequent images. The time interval between the images was about 4 s, which yields a satisfactory resolution of 15 tip locations for one rotation of the spiral. The intersection of these two contour lines was detected with an accuracy of about ±5 pixels corresponding to ±0.1 mm.

III. EXPERIMENTAL RESULTS

Without an electric field the tip of a single isolated spiral as prepared in our experiments describes a circle around a rotation center that is stationary in time (rigid rotation). Under the influence of a constant current between 0 and 30 mA the center of a single isolated spiral moves along a straight line with a constant velocity and at a constant angle with respect to the vector of the electric field. Both the velocity and the angle depend monotonical on the current. In our experiments the angle and the velocity of the drift at a given current remained constant within 10% for more than 1 h. Nevertheless, for the following experiments with the symmetric spiral pairs we restricted the observation time to about 50 min to minimize the effects of the aging process of the solution.

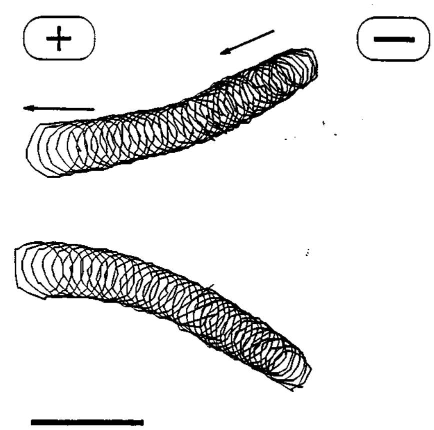

FIG. 2. Trajectories of the spiral wave tips observed at a constant current of 20 mA. The + and − signs denote the directions of the electric field. The arrows indicate the directions of the drift. The scale bar is 1 mm.

For values of the current exceeding 20 mA the counterrotating spirals approach each other and mutually annihilate as reported already by Schütz, Steinbock, and Müller [14]. In Fig. 2 the behavior for the spiral pair at a current of 20 mA is shown. Here the spirals approach each other for 30 min until the component of the velocity perpendicular to the electric-field vector disappears and they continue to drift parallel to the electric field in the direction towards the anode. In the experiments with lower current the observation time was too short to reduce the distance between the spirals sufficiently to observe the parallel drift. It was only possible to investigate the first part of the mutual approach.

Furthermore, we investigated the relaxation process of the spirals after switching off the current. In these experiments the distance between the spiral cores was reduced to less than one wavelength (approximately 1 mm) by applying a constant current of 30 mA for a relatively short time (10 min). Afterwards the current was switched off to investigate further interaction between the spirals. A relaxation process occurs in which the spirals repel each other and drift apart. This process is shown in Fig. 3 (long arrow).

IV. COMPUTATIONS

The experiments were complemented by numerical simulations of the observed phenomena. It is possible to modify the three-variable Oregonator model [16,17] to take into account the existence of an external field by introducing additional drift terms for the charged species [18,19] according to

$$\frac{\partial u}{\partial t} = \frac{1}{\epsilon}(qw - uw + u - u^2) + D_u \Delta u, \tag{1}$$

$$\frac{\partial v}{\partial t} = u - v + D_v \Delta v - K_v E \frac{\partial v}{\partial x}, \tag{2}$$

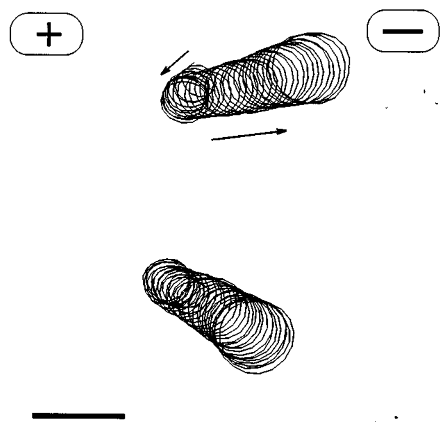

FIG. 3. Drifting trajectories of the spiral wave tips before (short arrow) and after (long arrow) switching off the current. The directions of the initial electric field is shown by the + and − signs. The scale bar is 0.4 mm.

$$\frac{\partial w}{\partial t}=\frac{1}{\epsilon'}(-qw-uw+fv)+D_w\Delta w+K_wE\frac{\partial w}{\partial x}. \quad (3)$$

The electric field influences the variable v (ferroin) and the variable w (Br^-), which are charged and have diffusion coefficients D_v, D_w different from zero. The variable u ($HBrO_2$) is not directly affected because it is not an ionic species. The ionic mobilities K_v and K_w are calculated from the charge and the diffusion coefficients to describe the additional fluxes $K_vE\partial v/\partial x$ and $K_wE\partial w/\partial x$ of both ions driven by the electric field E. D_u denotes the diffusion coefficient of the species u ($HBrO_2$). The numerical values of the parameters $q=0.002$, $f=1.4$, $\epsilon=0.01$, and $\epsilon'=0.0001$ as well as the values of the diffusion coefficients $D_u=1.0$ and $D_v=0.6$ were chosen according to Ref. [14]. The diffusion coefficient $D_w=1.12$ was calculated from the ratio of the molecular weights of $HBrO_2$ and Br^-.

Because of the large difference between the time scales of w and the other two variables u,v $(\epsilon'\ll\epsilon\ll 1)$, it is possible to assume that $w(x,y,t)$ is always determined by the instantaneous values of u and v according to

$$w=\frac{fv}{u+q}. \quad (4)$$

Due to Eq. (4), the dynamics of the variable u is closely coupled to that of w (anticorrelated), which is charged and therefore sensitive to the electric field. In the approximation of a small electric field one can reduce Eq. (1)–(3) to a two-component model

$$\frac{\partial u}{\partial t}=D_u\Delta u+\frac{1}{\epsilon}\left(u-u^2-fv\,\frac{u-q}{u+q}\right)+K_uE\frac{\partial u}{\partial x}, \quad (5)$$

$$\frac{\partial v}{\partial t}=D_v\Delta v+u-v-K_vE\frac{\partial v}{\partial x}. \quad (6)$$

These modified two-variable Oregonator model equations (5) and (6) take care of the field-induced behavior by including the effect of the electric field on the variable w in the equation for u by an additional flux term $K_uE\partial u/\partial x$. This term

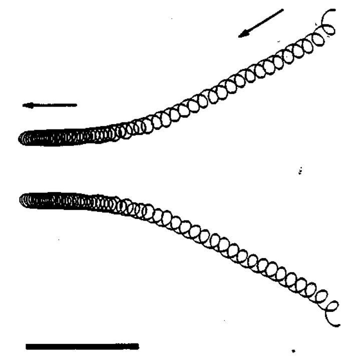

FIG. 4. Paths of the spiral tips for the computation with $E=0.2$. The direction of the drift is indicated by the arrow. The scale bar is 6.25 su (su=0.125).

does not describe any real physical flux of u, but results from the approximations made. The results presented in this paper are based on computations with this modified two-variable model only because the results from computations with the three-variable model turned out to be in satisfactory agreement with this reduced version. In our computations we studied the trajectories of the spiral wave tip for different values of the amplitude E ranging from $E=0$ to 0.24 and for different initial distances between the centers of the spiral cores. The computations were performed by an explicit Euler method using the nine-point approximation of the Laplacian and a symmetric approximation of the gradient term on a 384×384 array with a grid spacing $\Delta x=0.125$ and time steps $\Delta t=3\times 10^{-4}$. Pairs of spirals were generated by setting v to the steady-state value on the whole array and $u=0.7$ on a ten-grid-point-wide stripe on the left-hand side of the array and to the steady-state value elsewhere. After the planar wave had crossed two-thirds of the array, the upper and the lower part of the array were set to the steady-state values leading to a spiral pair evolving in the center of the array. These initial conditions were kept the same for all further computations.

V. COMPUTATIONAL RESULTS

With the chosen parameters it was possible to reproduce qualitatively the behavior that was observed in the experiments. Furthermore, we could investigate the dependence of the final distance of the spiral centers on the strength of the electric field in the case of the parallel drift. When setting $E=0$, the case without an electric field, the spirals do not interact if the distance of the centers is larger than about one wavelength. For values of the electric field exceeding $E=0.215$ we could observe a mutual approach of the spirals followed by an annihilation. The parallel drift was observed for values of the parameter E between these cases, as shown in Fig. 4 for a value of $E=0.2$. Remarkably, the final distance of the spiral cores depends linearly on the electric-field parameter (Fig. 5). Some details, however, differ in the computational results; in particular, one observes a slowing down of the drift in the course time, whereas the experiments in Fig. 2 indicate an acceleration of the drift velocity. This may be partially due to the aging effects of the closed system and also points to limitations due to simplifications in the model.

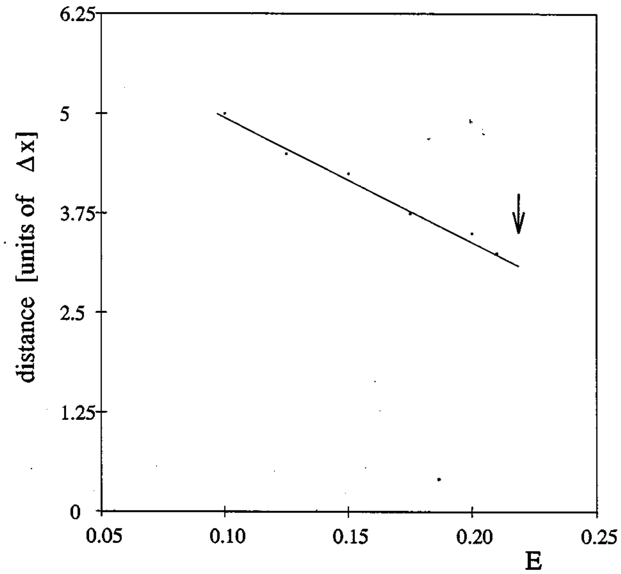

FIG. 5. Final distance of the spiral cores as a function of the parameter E. For values up to $E=0.215$ the dependence of the distance on E is clearly linear. Increasing E above $E=0.215$ (arrow) leads to a collision and annihilation of the spirals ($\Delta x = 0.125$).

We could also reproduce the relaxation process illustrated in Fig. 3. In this calculation we used the electric-field parameter $E=0.4$ to reduce the mutual distance of the spiral cores to less than one wavelength before resetting E to zero. The qualitative behavior as shown in Fig. 6 is the same as that observed in the experiments (Fig. 3).

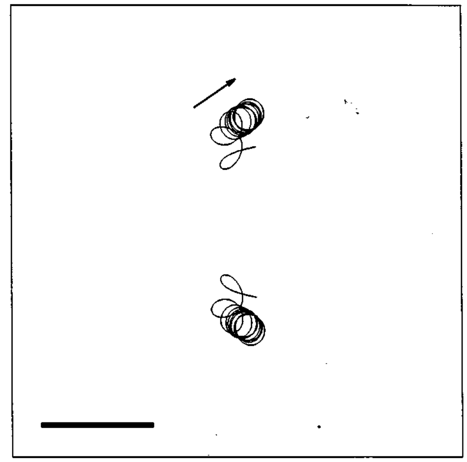

FIG. 6. Paths of the spiral tips for the relaxation. The arrow indicates the direction of the drift. The scale bar is 3.12 su (su $=0.125$).

VI. DISCUSSION AND CONCLUSIONS

The main result of the present work is the experimental observation of the parallel drift of two spiral waves under the influence of an external electric field. This effect was also reproduced in numerical simulations with a reaction-diffusion model. Our findings suggest that a repulsing force between spirals exist, which has a smooth dependence on the mutual distance of the cores until a minimal distance is reached at which the spirals annihilate each. This interaction of spiral waves in a symmetric arrangement is not only interesting in itself, but it can furthermore give insight into the problem of the behavior of spiral waves in the vicinity of no-flux boundaries of the medium because the case of symmetric spirals is equivalent to a single spiral close to a flat no-flux boundary. In fact, a parallel drift along a boundary has been shown in experiments of spiral rotation in a very small disk of agar gel loaded with BZ-reaction solution [20]. The advantage of using the interaction of a spiral pair is that it circumvents the problem involved in preparing a no-flux boundary in the gel, without introducing additional inhomogeneities. In our experiments, however, the use of a closed reactor turned out to be a disadvantage, because it did not allow us to extend the observation time enough to investigate the stability of the parallel drift or the dependence of the final distance of the spiral cores on the amplitude of the electric field in the case of the parallel drift. This calls for further experimental studies with an open reactor.

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