

## Electric-Field-Induced Drift and Deformation of Spiral Waves in an Excitable Medium

O. Steinbock, J. Schütze, and S. C. Müller

Max-Planck-Institut für Ernährungsphysiologie, Rheinlanddamm 201, D-4600 Dortmund 1, Federal Republic of Germany  
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The dynamic behavior of spiral-shaped excitation patterns in the Belousov-Zhabotinskii reaction was investigated under the influence of externally applied direct current. Spiral centers drift towards the anode. The chirality of the spirals determines the direction of an additional perpendicular drift. A deformation of the Archimedian spiral shape was observed. Both effects were studied quantitatively and were reproduced in simulations with a simple reaction-diffusion model.

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Spatiotemporal patterns formed in systems far from equilibrium have been investigated intensively in recent years. One of the most interesting and thoroughly studied systems remains the classical Belousov-Zhabotinskii (BZ) reaction [1], which is the oxidation of malonic acid by bromate in the presence of catalysts such as ferroin or cerium. Extended excitable systems such as thin layers of the BZ reaction exhibit patterns, such as rotating spirals or expanding trigger waves. In particular, the core region of spirals is a major focus of scientific interest [2]. For BZ media showing a sufficiently high excitability the core location remains stable in time; thus the spiral tip rotates around a circle. Remarkably, systems with low excitability show a different behavior: The spiral tip starts to meander [3], tracing out "floral" trajectories [4], but there are also experimental and numerical results showing more complex wandering of the tip along unpredictable traces [5]. Furthermore, Agladze, Davydov, and Mikhailov [6] and Mikhailov [7] suggested the possibility of controlled spiral drift by varying the excitability in time or space.

In this Letter we introduce a new technique with the goal of forcing spiral drift. This drift is caused by a small direct current, which influences the transport of ions. Sevcikova and Marek [8,9] studied this interaction in one-dimensional systems. They observed a speeding up of traveling waves towards the anode and a slowing down towards the cathode. Feeney, Schmidt, and Ortoleva [10] were the first who investigated the influence of electric fields on two-dimensional patterns in BZ media. These authors show that wave fronts can be broken or split by high electric currents.

Solution was prepared from distilled water and reagent grade chemicals and mixed with liquid agar (0.4%), which after solidification prevents hydrodynamic effects during the experiment. The final composition of the solution was  $0.05M$   $BrO_3^-$ ,  $0.05M$  malonic acid,  $0.2M$   $H_2SO_4$ , and  $6.25 \times 10^{-4}M$  ferroin [ $Fe(phen)_3SO_4$ ]. The mixture was placed in a flat, rectangular dish ( $71.6 \times 21.0$  mm<sup>2</sup>) with a layer thickness of 3 mm. Under these conditions patterns persist for more than 2 h. In order to account for the Ohmic heating ( $\sigma E^2$ ), a cooling box controlled the temperature of the dish at  $25 \pm 1.0^\circ C$ . A small glass-coated thermoelectric couple measured the temperature. The electrodes were placed parallel to the

short edges of the dish (Fig. 1). Metal brackets closed the electric circuit, which was driven by a dc power supply fixing the current to an arbitrary strength ( $I=0-40$  mA). The experimental pictures were taken by a charge-coupled-device video camera connected to a time lapse video recorder. For computational analysis the pictures were digitized with an image acquisition card ( $512 \times 512$  pixels, 8 bits gray level) and sent to a Convex-C201 computer.

Two different techniques contribute to the analysis of the experimental data. We first detected the traces of core drift by searching iteratively those pixel sites at which only very small gray level changes occur during one spiral period. This method takes advantage of the feature that the chemical activity inside the core region is very small [2]. The resulting coordinates of core location as a function of time were analyzed by linear regression. Second, we studied the spiral shape and its deformation by detecting that part of the wave profile which shows the strongest increase in intensity and calculating the corresponding polar coordinates  $(r, \phi)$  with respect to the core center.

The analysis of drifting spiral tips reveals a surprising behavior: The electric field forces the cores to drift towards the anode, with an additional component perpendicular to the field vector. The direction of this perpendicular drift depends on the chirality of the spirals, as

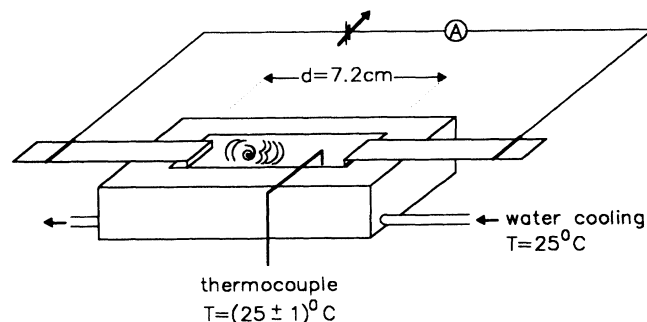


FIG. 1. Schematic representation of the experimental setup. Two  $K_2SO_4$  soaked paper strips are used as planar electrodes in order to apply an electric current on a thin agar layer carrying the BZ solution. The excitation patterns were detected by a video camera with a BG filter (Schott).

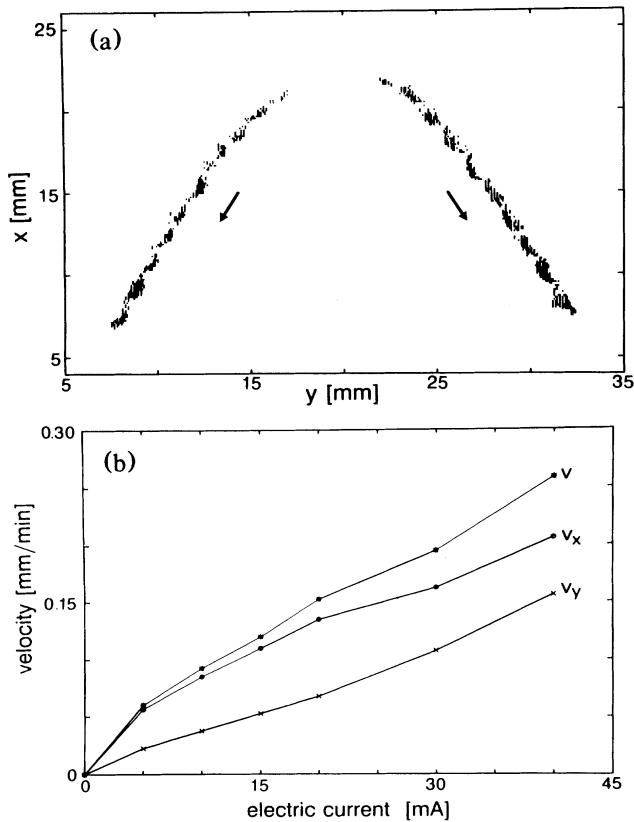


FIG. 2. (a) Core locations of two drifting spirals with different sense of rotation. Locations were estimated by detecting places of low-intensity changes. The applied electric current was  $I=35.0$  mA, with the anode at the bottom of the graph. (b) Dependence of velocity of drifting cores on current. The velocity component  $v_x$  is parallel to the electric field;  $v_y$  is perpendicular. The chirality of a spiral determines the sign of  $v_y$ .

shown in Fig. 2(a). A current of  $I=35.0$  mA, corresponding to approximately  $E=5$  V/cm, shifted the spirals to the anode at the lower part of the plot. The left spiral, rotating clockwise, possesses an additional drift to the left side. The other spiral, rotating counterclockwise, drifts towards the right side. Switching off the current immediately stops the core drift. Changing the polarity of the field causes a core drift towards the initial position.

The velocity and angle of the drift strongly depend on the current strength. Higher electric current yields faster drift and larger angles. This dependence is shown in Fig. 2(b). With  $0^\circ$  for the field direction, the drift angle for  $I=5$  mA is measured to be  $22.5^\circ$  and for  $I=40$  mA is measured to be  $37.2^\circ$ . Quantitative experiments using higher currents are difficult because the large heat production causes higher and uncontrolled excitability. The shape of the core traces is linear only to a first approximation. In the later stage of the process we usually observed smaller angles.

The shape of unperturbed spirals in this recipe is very

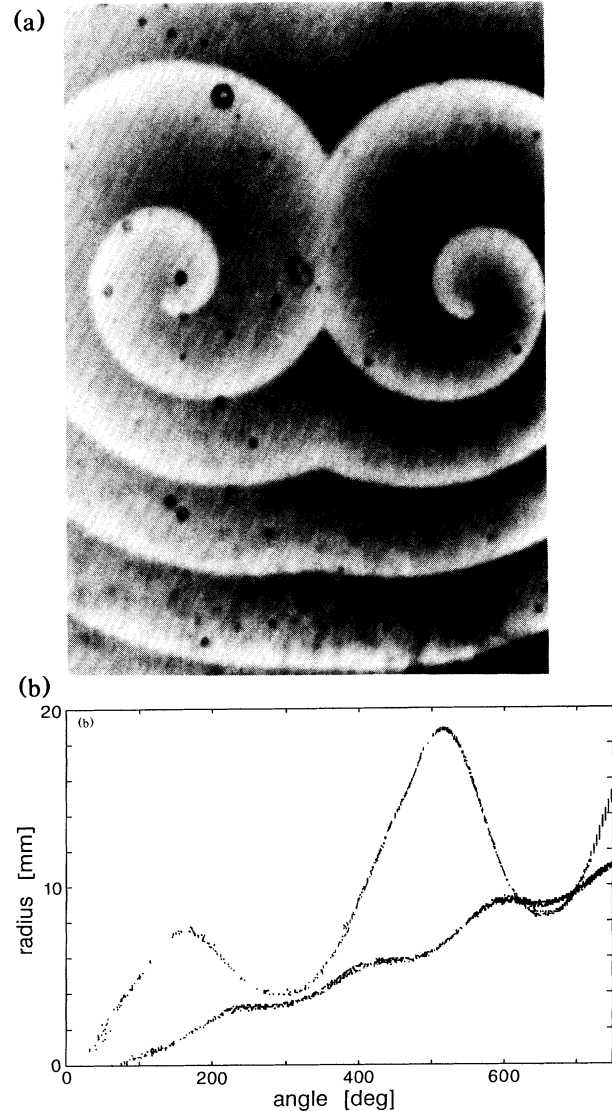


FIG. 3. (a) Image of deformed spiral patterns, with  $I=35$  mA. (b) Polar coordinates of two spiral wave fronts with respect to their core center. The unperturbed spiral ( $I=0$  mA) shows a linear (Archimedean) dependence. A current of 35.0 mA forces a deformation, which reaches its maximum in the back of the drift.

close to Archimedean [11]. A representation of its geometry in polar coordinates  $(r, \phi)$  leads to a linear function: Moving along one whorl of the spiral ( $360^\circ$ ) causes a displacement of one wavelength. Figure 3(a) shows spirals that are deformed by a 35.0-mA current. A  $(r, \phi)$  diagram of a deformed spiral is given in Fig. 3(b) including, for comparison, the shape without current. The deviation reaches its maximum in the back of the drift direction. The  $(r, \phi)$  coordinates of the deformed spiral arm approach the unperturbed state near the drift angle.

For strong currents we found "filaments" with low curvature traveling without shape change and highly curved

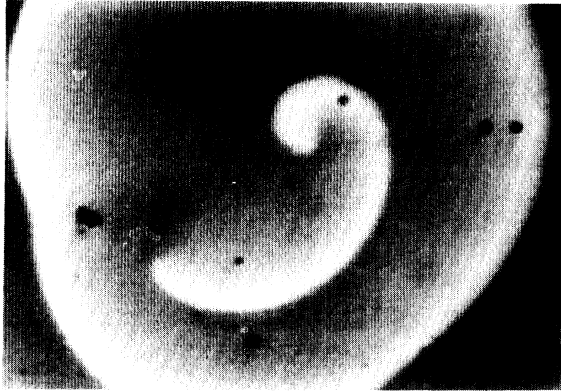


FIG. 4. Image of drifting spiral tips influenced by a 35-mA current. Most of the time the waves have the shape shown in the lower spiral, while cringing takes only a very short time (upper spiral).

wave ends that quickly cringe, as shown in the spiral pair of Fig. 4. The phase difference of rotation leads to an alternating occurrence of both types of structures at the two open ends. This phenomenon illustrates the main principle of forced spiral drift. In a first approximation, the action of the electric field on a wave front depends on the cosine of the angle  $\theta$  between field vector and normal wave velocity. The motion of the unperturbed spiral tip which is rotating around a circular core region with constant frequency causes a linear increase of this angle. The electric field modulates the rotation frequency  $\omega(\theta)$  and the absolute position of the tip, resulting in the observed behavior.

We suppose that the described phenomena are mainly due to a field-induced transport of the inhibitor  $\text{Br}^-$ . It should be noted that the temporal evolution of the bromide concentration is anticorrelated to that of the autocatalytic component  $\text{HBrO}_2$ . Effects of the external perturbation on the large ferroin complex might be less important.

A simple two-variable reaction-diffusion model can give insights into the mechanism of electric-field-induced drift and deformation, when the equation of the propagator variable  $u$  is modified by an additional convection term, as described by Schmidt and Ortoleva [12]:

$$\frac{\partial u}{\partial t} = D\Delta u + f(u, v) + ME \frac{\partial u}{\partial x}, \quad (1)$$

$$\frac{\partial v}{\partial t} = g(u, v), \quad (2)$$

with the diffusion coefficient  $D$ , the ion motility  $M$ , and the electric-field strength  $E$ . We used the nonlinear kinetic rate laws for excitable media suggested by Barkley [13].

By integrating this system of partial differential equations we found stable core positions for spirals without electric current. Nonzero values of  $E$  result in drifting

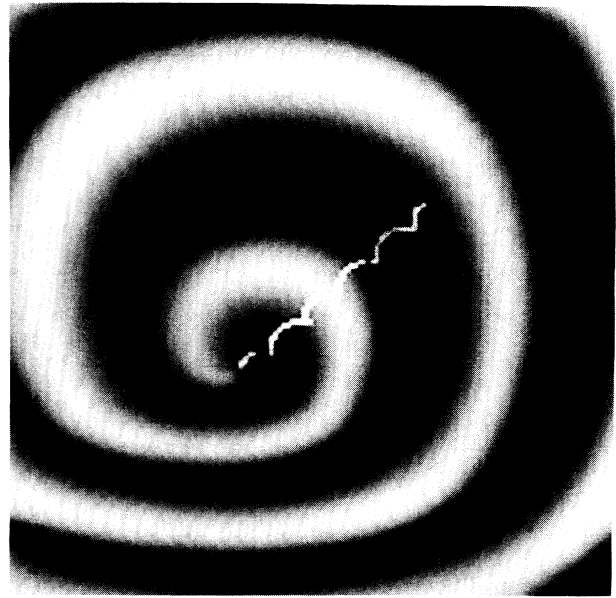


FIG. 5. Numerical simulation of a drifting and deformed spiral. The trace of the spiral core (bright dots) is superposed on the final spatial distribution of the variable  $v$ . In this example the value of  $ME/D$  was 0.35. Simulations were done by using a  $150 \times 150$  grid point array.

spiral centers and deformation of the spiral shape. A typical example of both effects is shown in Fig. 5. In this simulation the value of  $ME/D$  was 0.35. The evolution of the intersection of the  $u = 0.50 \pm 0.02$  and  $v = 0.34 \pm 0.03$  level curves provided the trajectories of drifting spiral tips.

Detailed analysis of drift velocities reveals a monotonic dependence on the electric-field strength  $E$  [Fig. 6(a)]. The velocity component parallel to the field  $v_x$  and the corresponding drift angle show smaller increases for larger values of  $E$ . The angle seems to converge to approximately  $60^\circ$ . Figure 6(b) presents the polar coordinates of spiral wave fronts without convection and for  $ME/D = 0.35$ . Without any electric field, we found only small wiggles in an overall linear dependence caused by the quadratic geometry of the grid cells and the simple five-point calculation of the Laplacian. For strong electric fields, there are significant deviations from the Archimedean shape, as found in the experiment.

All computational results are in good qualitative agreement with the experimental observations. Furthermore, such numerical simulations help to study the effects of stronger current under controlled conditions. The simplicity of the model used indicates that the phenomenon of drifting spirals is a common property of excitable media in which long distance transport takes place.

Numerical simulations reveal that only waves propagating into a completely recovered region show a linear dependence of wave velocity on electric-field strength. Drift and deformation appear to be strongly influenced by

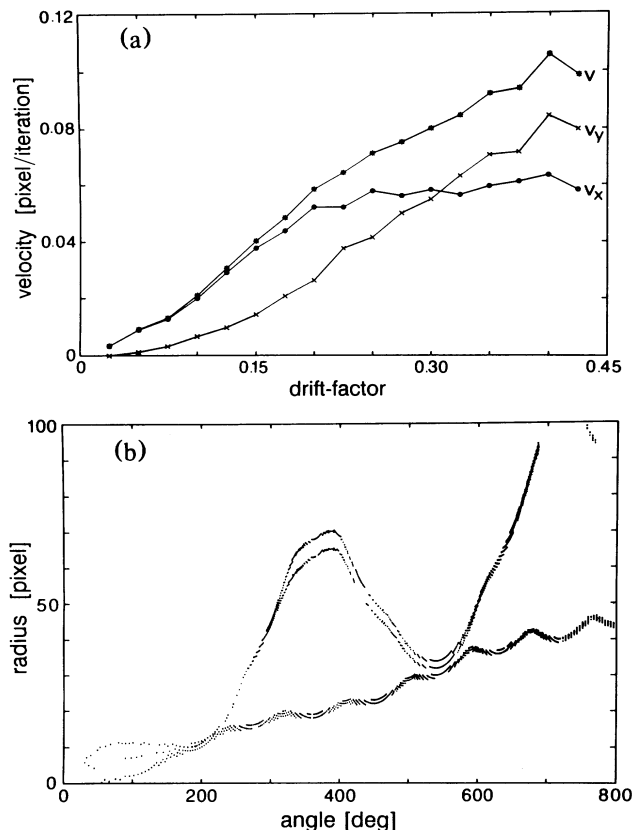


FIG. 6. (a) Numerically simulated drift velocities  $v$  as a function of drift factor  $ME/D$ .  $v_x$  describes the velocity component in the  $x$  direction (direction of the electric field);  $v_y$  is the perpendicular component. (b) Polar coordinates of two simulated spiral waves with respect to their core center. Increasing the value of  $ME/D$  leads to a strong deformation of the unperturbed spiral shape (compare Fig. 3).

the dispersion relation [14] of the system and by the characteristic action of the electric field.

We recently have obtained direct experimental evidence for the annihilation of spiral pairs that collide under the influence of electric fields. Furthermore, we observed a temporal interaction of spiral centers trying to

surround each other in order to continue their own unperturbed drift [15]. The experimental and computational techniques presented are important new tools for the field of pattern dynamics in excitable systems. These results will probably simplify the explanation of compound tip motion and will reveal new insights in the structure and stability of spiral cores.

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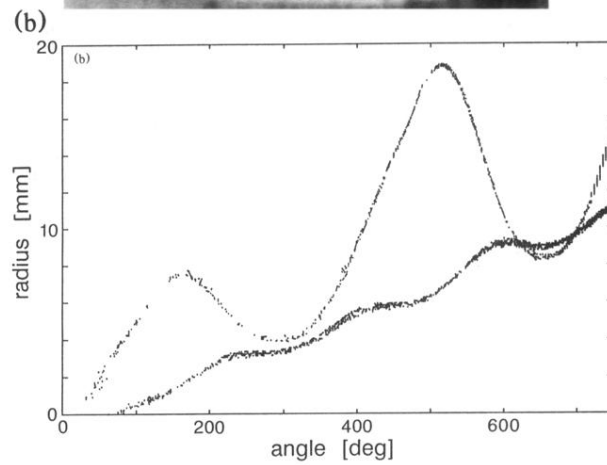
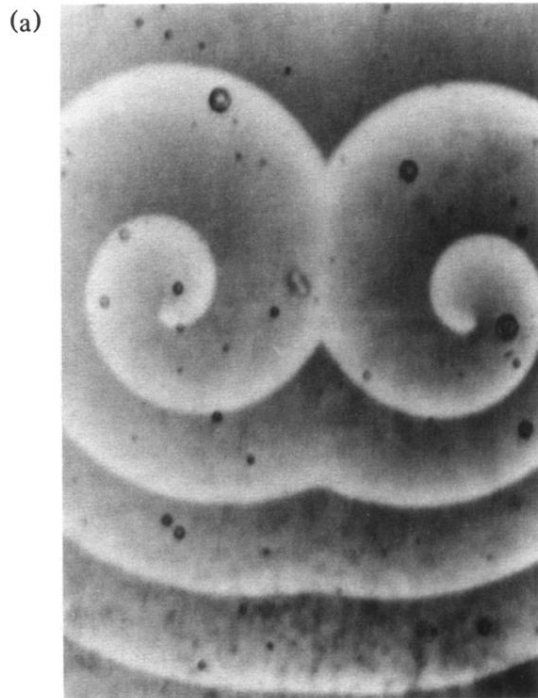


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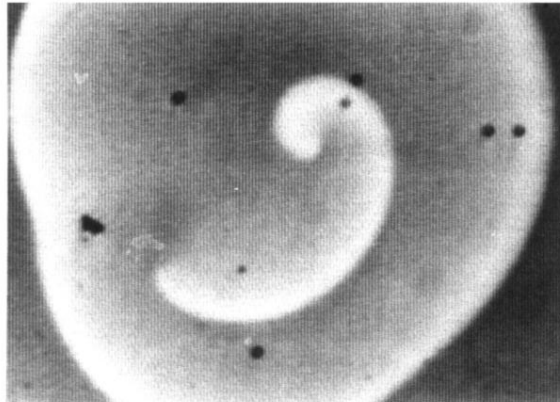


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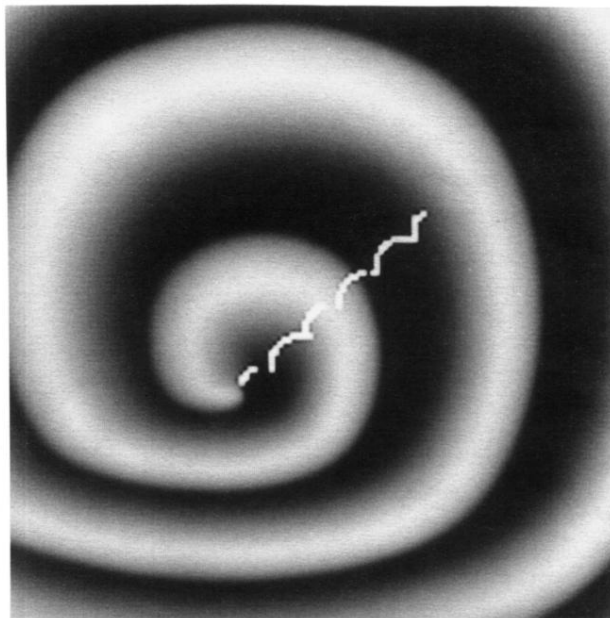


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