Interacting spiral waves in the Oregonator model of the light-sensitive Belousov-Zhabotinskii reaction

I. Schebesch and H. Engel
Institut für Theoretische Physik, Technische Universität Berlin, Hardenbergstraße 36, 10623 Berlin, Germany
(Received 1 June 1999)

We study the interaction of meandering spiral waves within the framework of a modified Oregonator model for the light-sensitive Belousov-Zhabotinskii medium. In this medium the local excitation threshold can be controlled by varying the intensity of incident light. At low as well as sufficiently high light intensity we find stable axis-symmetric bound states consisting of two counterrotating spirals. At intensity values in between, spiral pairs undergo a symmetry-breaking instability, leading to one spiral suppressing and expelling the other. To avoid the instability, we consider a spiral wave interacting with its mirror image close to a plane boundary impermeable to diffusion. The drift velocity and the drift direction of those pseudobound states parallel to the boundary are strongly influenced by the light intensity. [S1063-651X(99)04012-X]

PACS number(s): 05.65.+b, 05.45.−a, 47.54.+r, 82.20.Wt

I. INTRODUCTION

The dynamics of isolated spiral waves has been studied in systems of quite different nature. The spectrum covers rotating waves of chemical activity in the Belousov-Zhabotinskii (BZ) reaction [1], coverage patterns of adsorbed species on platinum single crystal surfaces during CO oxidation under ultrahigh vacuum conditions [2], cAMP waves in aggregating social amoeba colonies such as the slime mold *Dictyostelium discoideum* [3], circulating waves of neuromuscular activity in cardiac muscle tissue [4], spiral waves of intracellular calcium release [5], and many others. Frequency selection, annihilation of colliding wave fronts, and trapping at inhomogeneities are phenomena that occur generically in all the mentioned active media. It is well known that spiral wave dynamics can be complicated ranging from one frequency, simple rotation to quasiperiodic, compound rotation, and to spiral turbulence [6].

Comparatively less is known about the interaction of spiral waves. Experimentally one finds that the waves emitted by the spiral source provide an effective screening for the influence of other spirals. Thus, a possible interaction is expected to be short ranged. At a distance between the cores of a few wavelengths it is extremely weak and negligible on any realistic time scale.

In a series of papers Aranson *et al.* [7] considered the problem of spiral wave interaction in the framework of the complex Ginzburg-Landau equation (CGLE). This equation describes the medium close to a supercritical Hopf bifurcation. It was found that the interaction between spiral waves decays exponentially at large distances of the spiral cores. At smaller separation, the interaction results in a relative motion of the spiral cores. The velocity of this motion possesses a radial component v_r, acting along the connecting line of the core centers, and perpendicular to it a tangential component v_t.

Depending on the parameters of the medium, the character of the interaction is attractive ($v_r<0$) or repulsive ($v_r>0$). In a narrow parameter range at small separation bound states can exist ($v_r=0$). A bound state formed by two counterrotating spiral waves which have opposite topological charge is axis symmetric. In this case the tangential velocity components are equal and point in the same direction ($v_{t1}=v_{t2}$). Thus, axis-symmetric bound states drift as a whole with constant velocity parallel to the symmetry axis. Two spiral waves with equal topological charge eventually form a bound state that has central symmetry. Then the direction of the tangential components is opposite ($v_{t1}=-v_{t2}$), and the spiral pair as a whole rotates with constant angular velocity around the common center of symmetry. For both types of bound states, the stationary distance between the spiral cores is of order of the wave length.

Regarding the stability of bound states, in the CGLE model a spiral pair undergoes a symmetry-breaking instability: one member of the spiral pair overwhelms its neighbor and pushes it to the periphery. This instability is related to a positive feedback of a random increase in the rotation frequency of one spiral [7].

For excitable media, early numerical studies performed by Ermakova *et al.* in 1989 with the FitzHugh-Nagumo model also led to the conclusion that bound states of spiral waves can exist [8]. Recently, the problem of spiral competition in excitable media was considered based on numerical simulations of a three-variable reaction-diffusion model [9]. Again, symmetry breaking in a spiral pair was found, leading to one spiral suppressing and expelling the other.

Experimental evidence for the symmetry-breaking spiral pair instability in light-sensitive BZ media has been found recently using an open gel reactor [10]. We take the chemical recipe used in these experiments as an orientation aid for the choice of appropriate simulation parameters. Within the reactor also the drift of a spiral wave parallel to a plane boundary impermeable to diffusion has been measured. For symmetry reasons, close to the boundary the spiral wave, interacting with its virtual mirror image, models an axis-symmetric spiral pair. Further experimental and computational results were reported in Ref. [11].

The numerical investigations in this paper focus on the influence of the excitability on the behavior of axis-symmetric bound states. The calculations are carried out for light-sensitive BZ media, whose local excitation threshold in

first approximation is proportional to the intensity of incident light. The BZ medium is described by a modified three variable Oregonator model. We show that varying the intensity of externally applied illumination we can control the stability and the dynamics of the spiral pairs. The parameter range investigated covers with increasing light intensity outward meandering, inward meandering, and rigid rotation at large cores. Being oscillatory for small light intensity the local dynamics becomes excitable, when a certain threshold is exceeded.

In Sec. II we briefly discuss the modified Oregonator model. The results of the numerical simulations are presented in Sec. III. We conclude with a short summary and discussion.

II. THE MODEL

In 1974 Field and Noyes proposed a minimal model for the BZ reaction based on five irreversible reactions among three chemical species: bromous acid, $HBrO_2$ (the autocatalytic variable), bromide ions, Br^- (playing the role of an inhibitor), and the oxidized form of the metal ion catalyst, M (ox) (the recovery variable) [12]. This kinetic description known as the three component Oregonator model can explain many features of the BZ reaction.

In the light-sensitive variant of the BZ reaction the ruthenium-bipyridyl complex is used as catalyst. In the usual scheme of the reaction, this complex promotes the autocatalytic production of the activator $HBrO_2$ only in its reduced and photochemically unexcited state. Once the ruthenium complex becomes photochemically excited, it slowly catalyzes the production of the inhibitor bromide. Thus, externally applied illumination can create an additional source of inhibitor Br^- and suppress the excitability of the medium. To take account of photochemically produced Br^- Krug *et al.* introduced an additional flow term into the equation for the bromide balance. The modified three variable version of the Oregonator model reads [13]

$$\varepsilon \frac{\partial u}{\partial t} = u(1-u) - w(u-q) + D_u \Delta u,$$

$$\frac{\partial v}{\partial t} = u - v, \tag{1}$$

$$\varepsilon \frac{\partial w}{\partial t} = \Phi + f v - w(u+q) + D_w \Delta w,$$

with the dimensionless variables $u=(2k_4/k_3A)U$, $w=[k_4k_5B/(k_3A)^2]W$, $v=(k_2/k_3A)V$, $\varepsilon=k_5B/k_3A$, $\varepsilon'=2k_4k_5B/k_2k_3A$, $q=2k_1k_4/k_2k_3$, $t=k_5B$ time, $x=(D_u/k_5B)^{-1/2}$ space, and $A=[BrO_3^-]$, $B=$[bromomalonic acid]+[malonic acid], $P=[HOBr]$, $U=[HBrO_2]$, $V=[M\ (ox)]$, $W=[Br]$. $k_1,\ldots,k_5$ are the rate constants in the Oregonator model. The parameter f is related to the stoichiometric coefficient h in the reaction step of the Oregonator scheme that describes the Br^- release by oxidation of malonic and bromomalonic acid ($f=2h$). The Laplacians Δw and Δu describe the diffusion of $HBrO_2$ and Br^-. The ratio of the diffusion coefficients D_w/D_u can be estimated from the molecular weights of the two species. Diffusion of v is omitted, because usually in experiments the catalyst is immobilized in a silicia hydrogel matrix to avoid disturbances of hydrodynamic origin in the reaction zone. The parameter ϕ describes the photochemically produced bromide flow. In first approximation it is proportional to the intensity of the incident light. The chemical recipe that was used in the experiments by Brandtstädter *et al.* [10] corresponds to the following parameters: $f=1.8$, $1/\varepsilon=8$, $1/\varepsilon'=720$, $q=0.002$, $D_u=1.0$, and $D_w=1.12$. We keep these parameter fixed during most of the calculations. Note, that for a fixed value of f, the parameter ϕ controls whether the system is in the oscillatory or the excitable regime. The chosen parameters with large values of the parameter ϕ yield a system in the excitable regime with high excitation threshold. A decrease of ϕ causes a transition into the oscillatory regime via a supercritical Hopf bifurcation at $\phi_{\mathrm{Hopf}}=0.001737$.

We have integrated Eqs. (1) numerically with no flux boundary conditions substituting time derivatives by finite differences according to an explicit Euler method and calculating the Laplacians by a five-point discretization. To keep the influence of the boundaries small the calculations are carried out on 800×800 arrays (160×160 Oregonator space units) with grid spacing $x=0.2$. As initial condition we take a symmetric configuration of two counterrotating spirals with wave lengths λ where the distance between the core centers was chosen to be $d\approx\frac{2}{3}\lambda$ to guarantee an interaction between the spiral waves.

III. RESULTS

A. Stability of bound states

Axis-symmetric bound states are denoted stable if the mean distance between the spiral tips, d_{tip}, after some transient approaches a constant value in the range of the spiral's wavelength as time goes to infinity. A bound state becomes unstable if the mean distance between the tips is growing in time.

To investigate the stability of the bound state with respect to changes in the excitability of the medium the parameter ϕ is gradually increased. Remember, that ϕ determines the excitation threshold and in first approximation is proportional to the intensity of incident light. Starting with $\phi=0$ we find the system in the oscillatory regime. Interacting spiral waves perform outward meandering at large cores. As an example, Figs. 1(a) and 1(b) depict the tip trajectories of both spirals, and the variation of the distance d_{tip} between the tips with time, respectively. $d_{\mathrm{tip}}(t)$ is a high amplitude oscillation with large period T that is caused by the large dimensions of the tip paths. The high frequency modulations in $d_{\mathrm{tip}}(t)$ belong to the motion along the petals of the meander pattern. As a whole the bound state drifts with constant velocity parallel to its symmetry axis. Asymptotically, the mean distance between the tips is constant.

While increasing the value of ϕ, the bound state remains stable until ϕ reaches a certain threshold ϕ_1. For the chosen parameters we have $\phi_1=0.0009$. At this point the spiral pair undergoes a symmetry-breaking instability: A sudden, initially small phase shift between the two spirals increases with time, and finally one member of the spiral pair overwhelms its neighbor and pushes it to the periphery. The se-

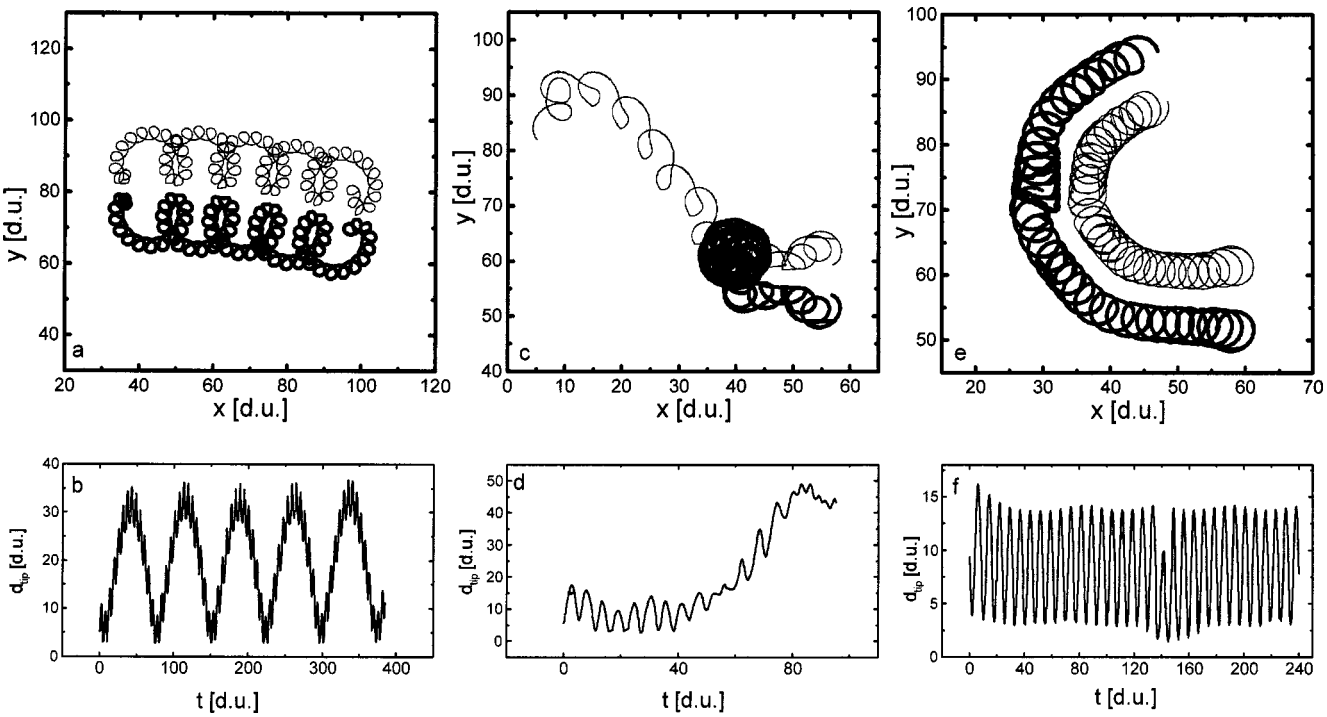

FIG. 1. Dynamics of axis-symmetric bound states in the modified Oregonator model for different excitability of the medium. (a), (c), (e) trajectories of the spiral tip, (b), (d), (f) time dependence of the distance between the tips of the interacting spiral waves. Parameter values: $f=1.8$, $1/\varepsilon=8$, $1/\varepsilon'=720$, $q=0.002$, $D_w/D_u=1.12$ (these values are the same for all figures, except Fig. 7). ϕ parameter: (a), (b) $\phi=0$, stable bound state; (c), (d) $\phi=0.002$, unstable bound state; (e), (f) $\phi=0.00375$, stable bound state near the stability boundary ϕ_2. d.u. denotes dimensionless Oregonator units.

quence in Fig. 2 captures this situation showing the time evolution of a symmetrical but unstable initial configuration. The corresponding tip trajectories, and the variation of the distance between them with time are shown in Figs. 1(c) and 1(d), respectively. Spiral pairs are unstable in a whole parameter range between ϕ_1 and a second threshold ϕ_2 which for the chosen parameters is $\phi_2=0.0036$. The transition back into the regime of stable bound states takes place at comparatively large values of ϕ where the tips of isolated, noninteracting spiral waves follow a periodic circular orbit of large diameter. The instability area covers a part of the oscillatory regime as well as a larger part of the excitable regime. These results are summarized in the stability diagram, Fig. 3. For completeness the figure also shows the tip path patterns of a isolated spirals wave under the same conditions. Note, that the symmetry-breaking instability of spiral pairs occurs close to the parameter value where single spiral waves undergo a transition from outward to inward meandering. Due to large space grids and simulation time it is difficult to decide whether these two thresholds actually coincide or not.

Let t_{delay} denote the time interval (measured in rotation periods T of an isolated spiral wave at the same parameters) that passes until the symmetrical bound state gains a phase shift obvious to the eye that increases and finally leads to the breakdown of the spiral pair. The dependence of the delay time on the parameter ϕ is plotted in Fig. 4. At the lower

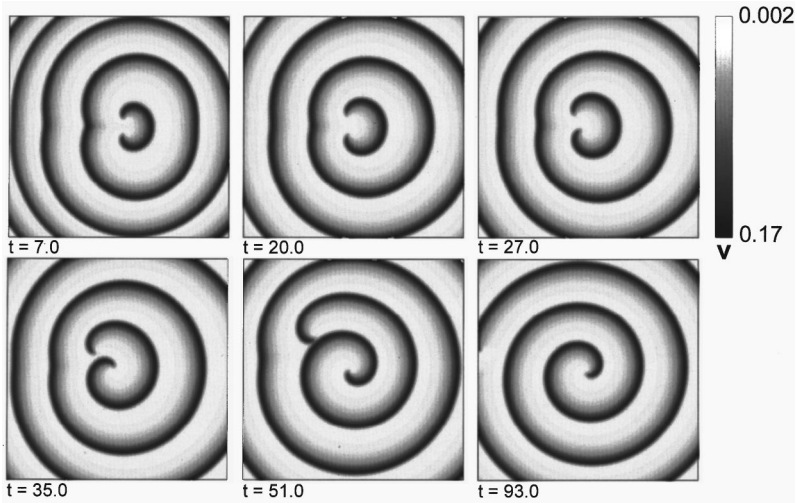

FIG. 2. Time evolution of two interacting spiral waves in the unstable parameter range between ϕ_1 and ϕ_2 ($\phi=0.0025$) starting with a symmetric initial configuration.

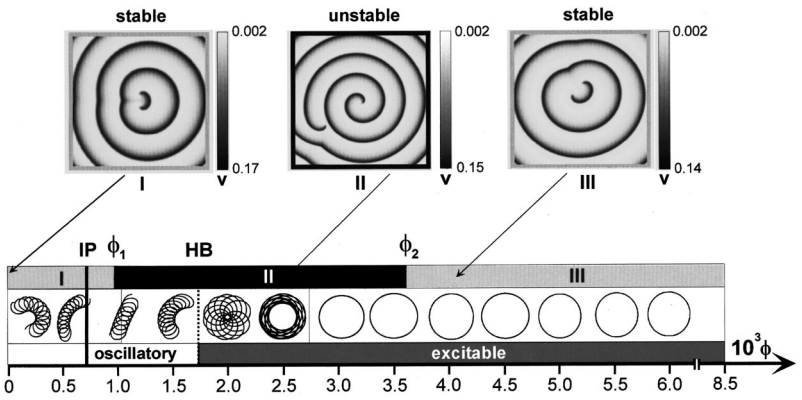

FIG. 3. Stability diagram of spiral pairs for varying intensity of incident light. Two interacting spiral waves form stable bound states in parameter region I and III. The unstable region is labeled II. Tip path patterns of single spiral waves (not in scale) are shown for illustration. For completeness the local dynamics is indicated too. HB denotes the supercritical Hopf bifurcation of the local dynamics. Spiral pairs change drift direction at ϕ value labeled IP.

boundary of the unstable area t_{delay} is quite large. With increasing ϕ it becomes smaller, and after the transition into the excitable regime it remains nearly constant up to the upper boundary of the unstable area. This dependence suggests different mechanisms for the destabilization of the bound state.

In the oscillatory regime we find a steep increase in the delay time when the stability boundary ϕ_1 is approached. This involves a large increase of simulation time. To determine an accurate as possible value for ϕ_1 we proceed as follows. At a ϕ value where an extrapolation of the graph from Fig. 4 predicts a delay time larger than $100T$, we follow the evolution of the spiral pair over a maximum of 200 rotation periods. When the spiral pair remains stable, we introduce an external phase difference increasing the parameter ϕ for a short time in the neighborhood of one spiral core. If this phase difference grows up and the bound state becomes unstable, we repeat the same procedure at a slightly smaller ϕ value. The boundary ϕ_1 corresponds to that value at which the artificially introduced phase difference persists or even decreases over more than 100 rotation periods without destabilizing the bound state. We remark, that neither an exponential function nor a power law give a satisfactory description of the increase in the delay time near ϕ_1.

At the stability boundary in the excitable regime ϕ_2, the delay times are relatively small. Close to ϕ_2 an initially symmetrical spiral pair always gains a phase difference after a few rotation periods and becomes asymmetric. Below threshold the asymmetry increases rapidly, and finally one of the two spirals is pushed to the periphery. Above threshold, however, this phase difference does not cause the destabilization of the bound state. Even when the phase difference first increases, later it may decrease again. The asymmetry remains small and changes with time leading to a spiral pair that moves through the medium by varying its direction of drift as shown in Fig. 5. The corresponding tip trajectories are plotted in Fig. 1(e). During more than 100 rotation periods the described behavior has been repeated several times without increasing the mean distance between the spiral tips [compare Fig. 1(f)]. Being pronounced in a narrow region close to ϕ_2, this scenario persists in weakened form for larger ϕ values. Far beyond ϕ_2 the stable spiral pairs are again axis-symmetric.

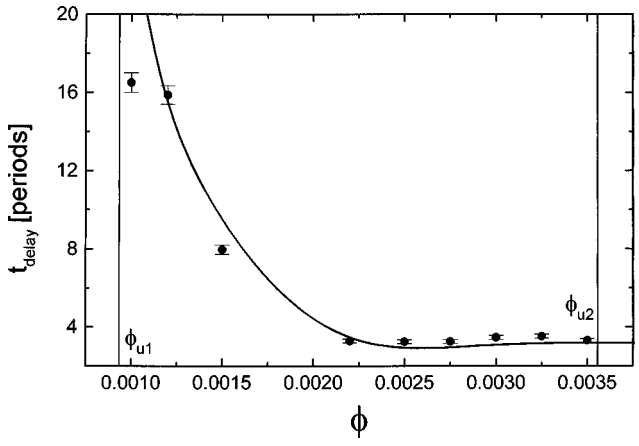

FIG. 4. Characteristic time for the breakdown of a spiral pair as a function of ϕ.

B. Drift of bound states

Stable axis-symmetric bound states formed by two counterrotating spiral waves drift as a whole with constant velocity parallel to their symmetry axis. To determine the drift velocity, we store the tip coordinates of both spirals at every 5 time steps. From the recorded data we obtain the positions of the core centers and determine the mean drift velocity of the spiral pair.

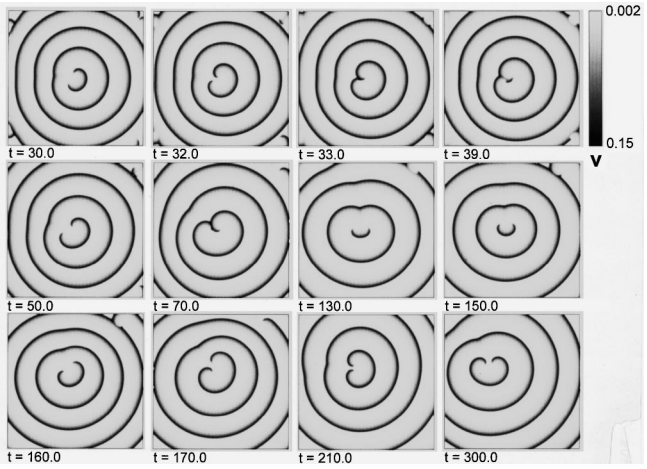

FIG. 5. Time sequence showing the dynamics of a bound state near the boundary ϕ_2 ($\phi = 0.00375$). A sudden phase shift decreases but causes a change in the drift direction. This process is repeated periodically without destabilizing the bound state.

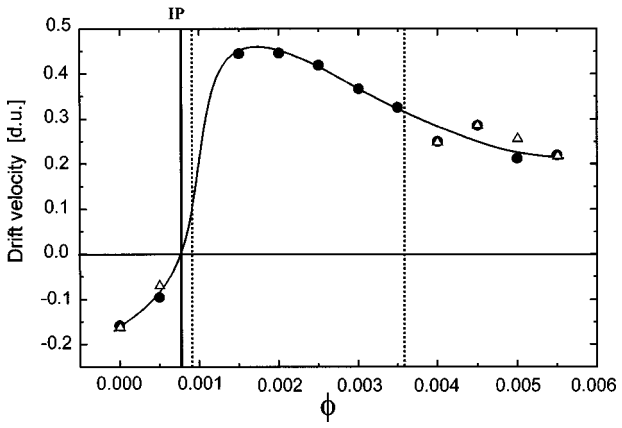

FIG. 6. Drift velocity of meandering spiral pairs for different values of light intensity. Open triangles: symmetrical spiral pairs in the area of stable bound states outside the dotted vertical lines. Filled circles: waves near the boundary (pseudobound states).

From the calculations follows that the drift velocity is usually one or two orders of magnitude smaller than the propagation velocity of the spiral wave. In the parameter region below ϕ_1, an increase of ϕ results in a slow down of the drift. At ϕ values larger ϕ_2, bound states drift at comparatively large velocities that decrease with increasing ϕ. Note, that stable spiral pairs below ϕ_1 and above ϕ_2 drift in opposite directions, compare Fig. 6, open triangles. The numerical determination of the point on the ϕ axis where the drift direction changes sign (labeled IP in Fig. 6) is very time-consuming as the inversion point is located close to the transition from inward to outward meandering for noninteracting spiral waves. Thus, the spirals are characterized by large wave lengths and perform compound rotation with large core size.

To obtain some information about the drift in the unstable area $\phi_1 < \phi < \phi_2$, we consider the interaction of a single spiral wave with its mirror image close to a plane boundary impermeable to diffusion (i.e., no flux boundary conditions).

To create such a pseudo-bound state, we shift a spiral wave to one of the boundaries. To feel the boundary the distance of the tip should be of the order of the wave length. Figure 7 shows typical tip trajectories of spiral waves propagating close to a plane boundary impermeable to diffusion. From those trajectories we again calculated those the mean drift velocity (filled circles in Fig. 6). In the parameter regions with stable bound states the results are in good agreement with the data obtained previously for axis-symmetric spiral pairs. The transition into the unstable area at low ϕ values is accompanied by a distinct increase in drift velocity. Then the drift velocity decreases again. By interpolating between the calculated values the inversion point IP was found close to the stability boundary ϕ_1.

IV. DISCUSSION

The presented results show strong influence of the excitation threshold on the stability of spiral pairs and on the velocity and the direction of their drift through the medium. Though the numerical calculations are carried out within the Oregonator model for the light-sensitive BZ medium, the main conclusions are expected to apply to other active media describing generic aspects of spiral pair dynamics independent of the details of the underlying reaction kinetics.

On the other hand, it is interesting to compare the numerical results with experimental data. The parameter values $f = 1.8$, $1/\varepsilon = 8$, $1/\varepsilon' = 720$, and $q = 0.002$ used during the numerical simulations correspond approximately to the experimental situation studied. A measure for the strength of the interaction is the ratio between the drift velocity of spiral pairs and the propagation velocity of the spiral wave. Depending on the parameter ϕ the numerically calculated value for this ratio is between 10^{-1} and 10^{-2}, in good agreement

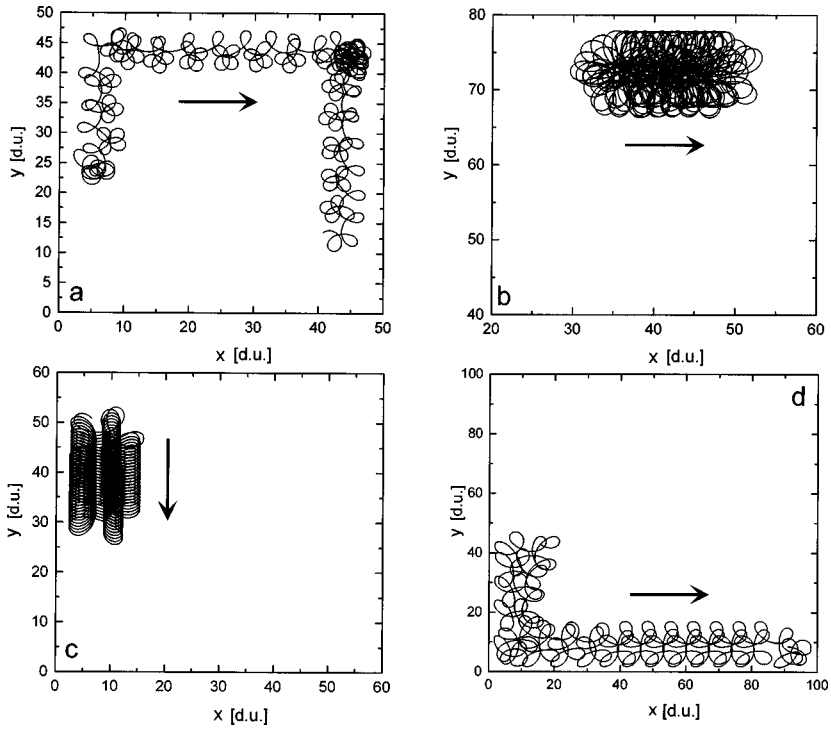

FIG. 7. Tip trajectories of meandering spiral waves propagating close to a plane boundary impermeable to diffusion for different values of ϕ. The arrows indicate the direction of the drift. Parameters: $f = 0.7$, $\varepsilon = 0.1$, $\varepsilon' = 0.22$, $q = 0.002$, $D_w/D_u = 1.12$. (a) $\phi = 0.004$, (b) $\phi = 0.008$, (c) $\phi = 0.009$, (d) $\phi = 0.01$.

with $c_{\mathrm{drift}}/c_{\mathrm{wave}} \approx 10^{-2}$ obtained experimentally with the same recipe [10]. Also, the numerically calculated values for the wavelengths between 1.5 and 3 mm, the rotation periods between 24 and 52 s, and the outer diameters of the tip path pattern between 0.6 and 2.8 mm are in the range of the experimental findings. A detailed quantitative comparison between numerical and natural experiment requires the relation between the intensity of incident light (in mW/cm) and the dimensionless photosensitive bromide flow ϕ and will be presented in a separate paper.

The results elucidate the relation between the local dynamics, the dynamic instabilities of noninteracting spiral waves and the dynamic behavior of spiral pairs. The parameter region where spiral pairs are unstable covers oscillatory as well as excitable local dynamics, and simple as well as compound rotation. Striking is how close to each other on the ϕ axis occur the breakdown of spiral pairs at ϕ_1, the transition of noninteracting spiral waves from inward to outward meandering, and the reversion of the drift direction of a single spiral parallel to a boundary impermeable to diffusion. In the numerical simulations there is always some uncertainty in the determination of the exact position of these boundaries. Thus, it remains still unclear whether they coincide or not.

We emphasize, that in the three-component Oregonator model the symmetry-breaking instability of spiral pairs occurs in a substantial parameter range. Remarkably, within a two-component version of the Oregonator model, obtained by eliminating the bromide balance adiabatically, we failed to reproduce the instability in the parameter range where the three-component model displays it. Moreover, to account for a possible shift of the stability boundaries Φ_1 and Φ_2 we have searched for the instability outside of this interval. Nevertheless, for simulation times up to 150 rotation periods we were not able to observe the instability. Similar observations were reported by Aranson *et al.* who showed that in contrast to the Ginzburg-Landau model a symmetric configuration of spirals in a two-component reaction-diffusion model is stable [14]. To our knowledge, the only counterexample has been reported by Ruiz-Villarreal *et al.* [11] who observed the instability in a two-component modified FitzHugh-Nagumo model simultaneously noting that the results depended on the grid mesh. Our results support the conjecture by Aranson *et al.* [9] that a third component is necessary to observe the symmetry-breaking instability of spiral pairs. However, the general prove, that only three-component models can do this instability, is still lacking.

ACKNOWLEDGMENTS

This work was partially supported by grants from the Deutsche Forschungsgemeinschaft and the Fonds der Chemischen Industrie.

[1] A. N. Zaikin and A. M. Zhabotinskii, Nature (London) **225**, 535 (1970); A. T. Winfree, Science **175**, 634 (1972); S. C. Müller, Th. Plesser, and B. Hess, Physica D **24**, 71 (1987); G. Li, Q. Quyang, V. Petrov, and H. L. Swinney, Phys. Rev. Lett. **77**, 2105 (1996).

[2] S. Jakubith, H. H. Rotermund, W. Engel, A. V. Oertzen, and G. Ertl, Phys. Rev. Lett. **65**, 3013 (1990); M. Eiswirth and G. Ertl, in *Chemical Waves and Patterns*, edited by R. Kapral and K. Showalter, (Kluwer Academic Publishers, Dordrecht, 1995), p. 447.

[3] G. Gerisch, Naturwissenschaften **58**, 420 (1983); K. J. Lee, E. C. Cox, and R. E. Goldstein, Phys. Rev. Lett. **76**, 1174 (1996).

[4] A. T. Winfree, *When Time Breaks Down* (Princeton University Press, New Jersey, 1987); J. M. Davidenko, A. M. Pertsov, R. Salomonsz, W. Baxter, and J. Jalife, Nature (London) **355**, 349 (1992).

[5] J. Lechleiter, S. Girad, E. Peralta, and D. Clapham, Science **252**, 123 (1991).

[6] W. Jahnke, W. E. Skaggs, and A. T. Winfree, J. Phys. Chem. **93**, 740 (1989); G. S. Skinner and H. L. Swinney, Physica D **48**, 1 (1991); M. Braune and H. Engel, Chem. Phys. Lett. **204**, 257 (1993); **211**, 534 (1993); D. Barkley, in *Chemical Waves and Patterns* [1], p. 163.

[7] I. Aranson, L. Kramer, and A. Weber, Physica D **53**, 376 (1991); Phys. Rev. E **47**, 3231 (1993); **48**, R9 (1993); Phys. Rev. Lett. **72**, 2316 (1994).

[8] E. A. Ermakova, A. M. Pertsov, and E. E. Shnol, Physica D **40**, 185 (1989).

[9] I. Aranson, H. Levine, and L. Tsimring, Phys. Rev. Lett. **76**, 1170 (1996).

[10] H. Brandtstädter, M. Braune, and H. Engel, in *A Perspective Look at Nonlinear Media*, edited by J. Parisi, S. C. Müller, and W. Zimmermann (Springer, Berlin, 1998), p. 271.

[11] M. Ruiz-Villarreal, M. Gómez-Gesteira, C. Souto, A. P. Munuzuri, and V. Pérez-Villar, Phys. Rev. E **54**, 2999 (1996); Phys. Rev. Lett. **78**, 779 (1997).

[12] R. J. Field and R. M. Noyes, J. Chem. Phys. **60**, 1877 (1974); J. J. Tyson, in *Oscillations and Traveling Waves in Chemical Systems*, edited by R. J. Field and M. Burger (Wiley, New York, 1985), p. 93.

[13] H. J. Krug, L. Pohlmann, and L. Kuhnert, J. Chem. Phys. **94**, 4862 (1990).

[14] I. Aranson, D. Kessler, and I. Mitkov, Phys. Rev. E **50**, R2395 (1994); Physica D **85**, 142 (1995).