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Long-term vortex interaction in active media

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The long-term interaction of two unlike spirals is studied in the framework of the Belousov-Zhabotinsky reaction. Both repulsive and attractive interactions have been observed for different initial distances between spirals. We describe how a spiral pair can evolve in three different ways; namely, spirals attract each other, collide, and annihilate; spirals repel each other and one of them dominates; and, finally, both spirals coexist without dominance of one of them. [S1063-651X(96)03809-3]

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Many extended systems exhibit long-range order caused by short-range interaction. Topological defects can appear in these kinds of systems and they determine to a certain extent macroscopical properties of real systems. Defects have been shown to play an important role in the onset to turbulence [1], since they can destroy long-range interaction in the media where they appear [2]. In oscillatory media, defects can take the form of spiral waves as in reaction-diffusion systems like the Belousov-Zhabotinsky (BZ) reaction [3–6] or some cases of catalytic surfaces [7]; or the form of dislocations as in anisotropic systems supporting traveling waves, e.g., electrohydrodynamic convection (EHC) in thin cells at not too low frequencies [8].

Although a lot of work has been devoted to the understanding of the interaction between spiral waves both theoretically and numerically [9–22], as far as we know there is no experimental evidence of most of the behaviors predicted by the previous approaches. This disagreement among theory, computation, and experiments is mainly due to two facts: namely, on the one hand, the sets of parameters considered in the theoretical approaches are scarcely reproducible in a laboratory; and, on the other hand, the computational approaches permit us to keep indefinitely the properties of the medium, what is difficult to achieve experimentally [a continuously fed tank reactor (CFTR) [23] must be used]; and, besides, discretization inaccuracies can dissipate long distance interaction.

Nevertheless, some authors have studied experimentally the interaction between spirals; therefore Agladze and Krinsky [24] and Steinbock and Müller [25] analyzed the existence of multiarmed spirals, Krinsky and Agladze [26] studied the interaction between these multiarmed spirals and spiral pairs, and Schütze *et al.* [27] considered forced vortex interaction and annihilation in an active medium (the BZ reaction).

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The goal of this Brief Report is to study the long-term interaction between a pair of unlike spirals in the framework of the BZ reaction. Our experiments were carried out in a BZ reaction, where the catalyst (ferroin) was immobilized in a silica gel [28] at room temperature ($25\pm1\,°C$). A 1-mm-thick gel was prepared in a Petri dish 88 mm in diameter, which was embedded in a bath where it remained covered by a thick liquid layer (2 cm) of the other BZ reagents [$NaBrO_3$ 0.17 M, H_2SO_4 0.17 M, and $CH_2(COOH)_2$ 0.17 M, which corresponds to an oscillatory medium]. In this way, interaction between the reaction and the oxygen in the air was prevented. Reagent properties were kept constant during the experiments—lasting at least 6 h—by imposing a flow of reagents into the bath (100 cm^3/h). Besides, the bath was homogeneously fed to avoid directional changes in chemical concentrations that could influence the movement of spirals.

Two unlike spirals were generated as follows: The medium was excited at a certain point by touching the gel with a silver wire [29] in order to generate a circular wave spreading through the medium from that point. Two discontinuous wave fronts were generated either by inhibiting a part of the front with a piece of iron [6] or by vulnerability [30]. These discontinuous wave fronts evolved into a pair of unlike spirals. They were created at the center of the medium to avoid boundary influence [31]. Note that, due to the generation method, both spirals do not have exactly mirror symmetry at the beginning of the experiment, although they have initially the same properties (wavelength $\lambda=0.30\pm0.01$ cm and period $T=140\pm2$ s). The experiments were followed with a charge-coupled device (CCD) camera connected to a Silicon Graphics workstation where images were digitized and spiral tip positions were automatically measured every 3 sec.

For different initial distances between spiral tips of initially slightly asymmetrical spirals, we have observed different types of interactions. If their initial separation is smaller than a certain critical distance ($d_c\approx2/3\lambda$, where λ is the

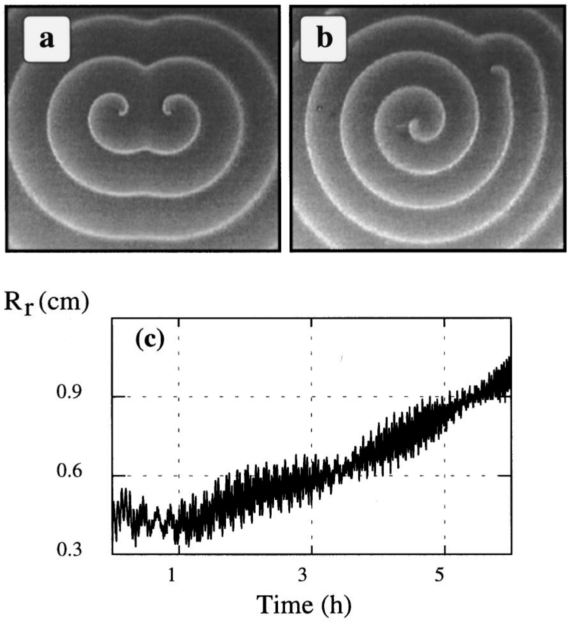

FIG. 1. Experimental evolution of two interacting spirals. A nearly symmetrical state some minutes after creating spirals (a) evolves so that spiral on the left (spiral 1) dominates and spiral on the right (spiral 2) is reduced to its bare core (b). The relative distance between spiral tips $[R_r = r_2(t) - r_1(t)]$ as a function of time is plotted in (c), where the repulsive character of interaction can be seen. The observed oscillations are due to the movement of each spiral around its core.

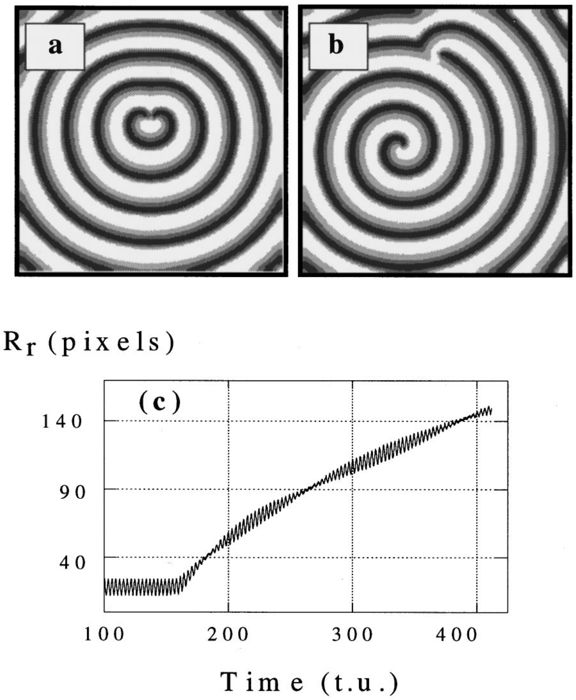

FIG. 2. Numerical simulation of two symmetrical interacting spirals. Using a modified FitzHugh-Nagumo model $[\dot{u} = \varepsilon(u - u^3 - g\nu + I) + D_u \Delta u; \dot{\nu} = u + b - k\nu]$, it is possible to observe the asymmetrization of initially symmetrical states like the one shown in (a). In (b) is shown the state of the system after 400 t.u., with one spiral developed and the other reduced to its bare core. The dependence of relative distance between spiral tips on time is plotted in (c). Parameters: $\varepsilon = 20$, $g = 1$, $I = 0$, $b = 0.6$, $k = 1$, $D_u = 5$, $dt = 10^{-3}$, $dx = 0.5$.

wavelength previously defined), interaction is attractive, that is, relative distance between spiral tips decreases until they annihilate each other. For initial distances of around one wavelength $(2/3\lambda \leqslant d \leqslant 5/4\lambda)$, spirals coexist but not in a stationary way, their relative distance and the angle between them (angle between the line connecting cores and the OX axis) oscillate in time. Above that distance, interaction is repulsive and causes spirals to move away from each other. The strength of the interaction decreases with initial distance; therefore, for large initial separations (several wavelengths), the tip movements of both spirals are only slightly affected by the interaction or, at least, the characteristic interaction time is much longer than that allowed experimentally.

Repulsion does not affect both spirals in the same way, but one of them starts dominating the other one as shown in Fig. 1. In Fig. 1(a) (30 min after starting the experiment) both spirals are almost symmetrical. In Fig. 1(b) (4 h later) one of the spirals has dominated and developed two wavelengths, while the other one has been reduced to its bare core. The time evolution of the relative distance between spiral tips is plotted in Fig. 1(c). During the first hour, one of the spirals starts dominating but the mean distance between their tips does not vary. After that transient, one of the spirals has reduced the other one to its bare core, and starts pushing it away in such a way that the relative distance increases. Note the existence of a short-term oscillation in the relative distance signal (sometimes this oscillation is around 0.2 cm), which is due to the size of both cores (their estimated radius is $R_c = 0.05$ cm) and to measuring inaccuracies. Besides, the oscillation shows some modulation due to the different rotation velocities of both spirals around their cores (the dominated spiral rotates slower). At first, in the symmetrical initial state, spirals are almost in phase, that is, each of the two spiral tips comes to the point of its core closest to the other spiral at the same time, and the same for the farthest one. The difference in velocities introduces a phase difference that grows during the experiment. As a result of this, spirals initially in phase (minimal oscillation amplitude) come to an out-of-phase state (maximal oscillation amplitude) and then to an in-phase state again and so on. It has been observed in several different experiments that the spatial distance between two minima corresponds to one wavelength.

Similar results were obtained in numerical simulations of two interacting spirals in a modified FitzHugh-Nagumo model [32] by means of a FTCS explicit scheme $(dt = 10^{-3}, dx = 0.5)$ with zero flux boundary conditions. Figures 2(a) and 2(b) illustrate how an initially symmetrical state ($\lambda = 25$ s.u., $T = 3.07$ t.u.) becomes asymmetrical. The dependence of relative distance on time is qualitatively the same in this numerical case [Fig. 2(c)] as in the experimental case [Fig. 1(c)]. It can also be seen in Fig. 2(c) that, after some iterations, the symmetry is spontaneously broken, and

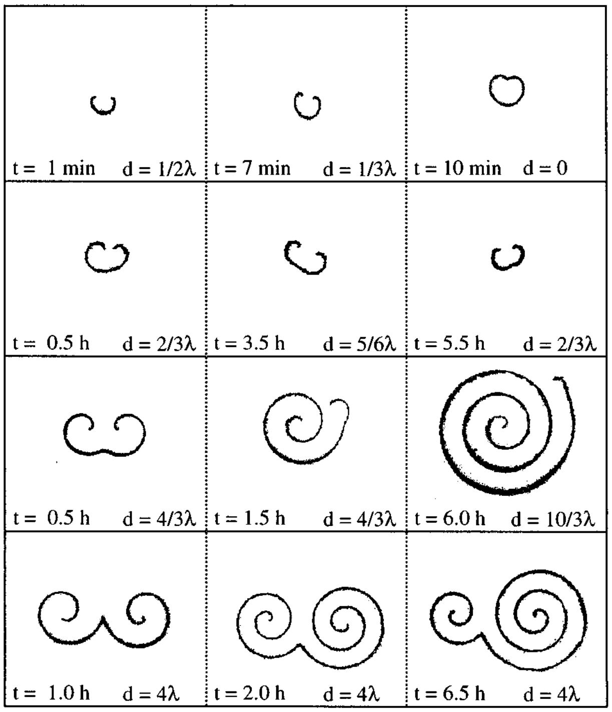

FIG. 3. Summary of the different types of interaction. Each row (three panels) represents the time evolution of a spiral pair for different initial distances between spiral tips—distances (in units of λ, wavelength) are averaged during one period. Note that only the curve connecting both tips is depicted; the continuous wave fronts formed after spiral collisions (similar to those observed in Fig. 1) were removed from the picture for the sake of clarity.

the relative distance starts growing with the same features described for the experimental case. In this case, spirals are created having perfect mirror symmetry and become asymmetrical due to numerical noise. It should be noted that results depend on grid mesh. We have observed that the smaller the space step, the longer the interaction range. Nevertheless, for a sufficiently small space step, the same qualitative behavior has been found.

Previously described behaviors are summarized in Fig. 3, where each row represents the time evolution of two almost symmetrical spirals whose tips were placed at a certain distance (only the curve connecting tips is depicted). As we have remarked, distance has a short-term oscillation; therefore, distances were averaged in one period. In the first row, the annihilation of two spirals initially at a distance $d \approx 1/2\lambda$ apart is shown. Note that instead of spirals, we should talk about ''protospirals,'' since the short distance between tips prevents spiral formation. In the second row, where $d \approx 2/3\lambda$, it can be seen how the relative distance oscillates in time, but neither annihilation nor domination is observed. In the third row, spirals initially placed at a distance $d \approx 4/3\lambda$ are observed to evolve so that one of them dominates and the other is reduced to its bare core, in such a way that the distance between spirals increases to more than three wavelengths during the experiment. Note that distance between tips is the same in the first two panels, since, during the first hour, one of the spirals grows and reduces the other to its core, whereas their positions only vary slightly. Finally, in the fourth row a similar situation is shown for an initial distance of $d \approx 4\lambda$, but, although one of the spirals dominates, the mean distance between spiral tips remains constant during the experiment. It can be observed how the shock (or sink) where both waves collide moves in the direction of the smaller frequency spiral.

Some phenomena related to spiral interaction have been widely treated in the literature. So, annihilation and coexistence are two well-known phenomena. Nevertheless, creation of asymmetrical states is quite an unusual phenomenon. Thus, Winfree [18] considered the existence of a bistable medium where two spirals with different rotation properties interacted. However, the relative distance between spirals did not vary in time but one of the spirals synchronized the other one in such a way that, finally, both oscillated with the same frequency. Other authors (e.g., Ermakova *et al.* [17] and Plesser and Müller [33]) considered numerically the existence of initially asymmetrical states, but they did not find this to influence the spiral pair dynamics—in [17] a final symmetrical state was reached and in [33] the asymmetry did not evolve in time. We believe this disagreement between numerical and experimental data is mainly due to two facts: namely, the interaction between spirals is a weak phenomenon, which needs in long-lasting experiments or numerical simulations to become apparent—in [17] the simulations lasted about 20 rotation periods and in our experiments and simulations we considered them to last more than 200 rotation periods. Besides, due to the weak nature of the effect, the spatial and time steps can strongly influence the tip's dynamics without apparent influence on the global behavior of both spirals—in our simulations we have observed that, above some critical distance, both spirals do not interact and that critical distance depends strongly on the grid mesh. Thus, in some way, the discrete nature of numerical simulations may influence the interaction dynamics, which is continuous in nature.

As far as we know, only Weber *et al.* [22], Bodenschatz *et al.* [16], and Aranson *et al.* [11] have observed how, in a spiral pair, one of them is able to dominate—the possibility of testing these computational results in the BZ reaction is proposed in [16]. They carried out numerical simulations of a generalized Ginzburg-Landau equation in an Eckhaus stable regime. In [16] they found the existence of some interval of initial distances inside which spirals interact in such a way that, after some transient, the up-down symmetry breaks and one of the spirals is able to dominate and push the other one away. In [11], the interaction is assumed to make spiral frequencies different, and so the larger frequency spiral will dominate. This difference in frequency was observed in our experiments averaging each frequency for several periods. However, we have not been able to determine differences in frequency from only a few periods (which would be analogous to an instantaneous frequency), because the difference between spiral periods is observed to be on the order of the time between consecutive determinations of spiral tip positions.

Finally, we have found that the initial distance is not the only parameter to describe spiral interaction; the symmetry of the initial pattern can strongly affect the final behavior.

Thus, when initially placed close to each other, the time in which spirals coexist before annihilation is not only determined by initial distance, but it is also found to depend on the symmetry of the initial pattern. For longer distances, the tendency of one of the spirals to dominate is accelerated by creating an initially asymmetrical pattern; note that in [16] the same effect is achieved adding noise. The influence of initial symmetry on the final state is in progress [34].

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