Drift of Interacting Asymmetrical Spiral Waves

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Long-term experiments on spiral interaction carried out in the framework of the Belousov-Zhabotinsky reaction have revealed the influence of the initial pattern symmetry on further pattern evolution. This symmetry is characterized by the different distance from each spiral tip to the boundary where the emitted wave fronts collide. The spiral initially closer to the shock line is observed to rotate with a lower mean frequency and to be dominated by the other one. Besides, both spiral tips are observed to drift. Different relationships between tangential and normal drift velocities of both tips have been found depending on whether domination is total or partial. [S0031-9007(97)02318-1]

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Spiral waves, one of the most interesting spatiotemporal structures that appear in reaction-diffusion systems, have been the subject of exhaustive studies during the last decades [1,2]. Despite the efforts devoted to understanding their dynamics [3–6], there exists no satisfactory description of them. Several studies [5,6] have stated that their properties are at some extent independent of the particular system (physical, chemical, or biological) where they are observed. The Belousov-Zhabotinsky (BZ) reaction [7] constitutes a fruitful tool for studying spiral wave dynamics, due to the relative simplicity of the experimental setup necessary to investigate their properties in different circumstances. It is known that once formed, the behavior of a spiral depends on the parameters of the medium [8]. However, the presence of other spirals [9], an external forcing [10], or the finite character of the medium [11] may break the translational symmetry in the medium and induce its displacement. Investigation of these phenomena can help to understand their dynamics and may provide mechanisms for controlling their behavior.

Long-term experiments in BZ reaction [9] have shown different types of evolution for two almost symmetrical unlike spirals in interaction, depending on the initial distance between their cores. If this distance is shorter than a certain critical value, spirals get closer and closer until they annihilate. For longer distances, one of them dominates after some time interval, which depends on the initial separation. This domination is illustrated in Fig. 1, where two spirals nearly symmetrical at first and separated a distance of 1.6λ evolve in such a way that one of them dominates and the other one is reduced to its bare core. Figure 1(a) shows the initial state (some minutes after starting the experiment), when both spirals are almost symmetrical. After the first hour, despite the mean relative distance between tips $\langle R_d \rangle$ has scarcely varied [see Fig. 1(d), where the time evolution of the relative distance between tips is plotted], one of the spirals has clearly dominated the other as shown in Fig. 1(b). This domination process makes one of the spirals to develop some wavelengths [in Fig. 1(c), the spiral on the left has developed two wavelengths] while the other one is reduced to its core.

In this paper, we show that the parameter which actually determines the behavior of two interacting spirals is not the distance between core centers, but the distance from each spiral core to the boundary where the fronts emitted by each spiral collide—we will call it shock line. We have observed that there always appears a drift movement superimposed to the rotation of each spiral around its core. The magnitude of interaction can be estimated by comparison between the modulus of the observed drift velocity with the linear rotation velocity of the spiral in the medium. Two unlike interacting spirals are observed to drift with velocities bigger when one of the spirals is totally reduced to its core than when they are initially almost symmetrical and far apart. The correlation between components of drift velocities is also different in both situations. In both cases, we deal with a small effect, which determines that it becomes apparent only after a long evolution.

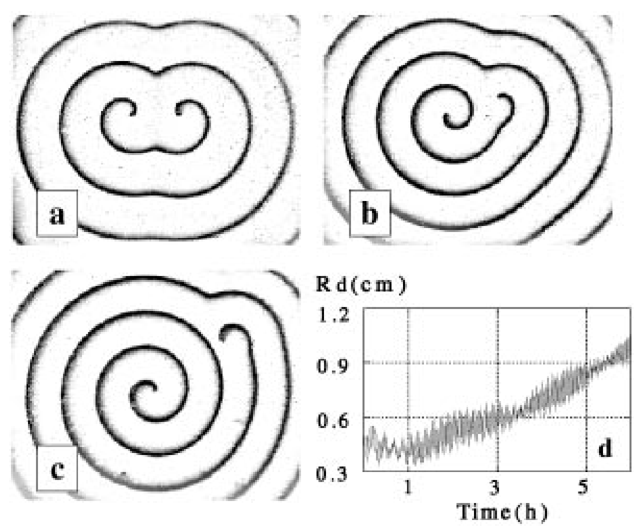

FIG. 1. Experimental evolution of two interacting spirals initially separated a distance 1.6λ ($\lambda \approx 0.3$ cm).

Our experimental setup constitutes an example of CFUR (*continuously fed unstirred tank reactor*) [12], which allows the observation of spiral waves in BZ reaction for long periods of time (our experiments last at least 6 hours, around 200 spiral periods). In our experiments, the catalyst (ferroin) was immobilized in a silica gel [13] at room temperature 25 °C. A 1 mm thick gel was prepared in a Petri dish 88 mm in diameter. This Petri 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 correspond 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 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 spiral movement. In fact, no drift of a single spiral was observed with this setup, and the results remained unchanged for different initial angles between spiral cores.

Two unlike spirals were generated as follows: The medium was excited at a certain point by touching the gel with a silver wire [14] 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 [15] or by vulnerability [16]. These discontinuous wave fronts evolved into a pair of unlike spirals (with the chosen concentrations, a single spiral in the medium presents a wavelength $\lambda = 0.30 \pm 0.01$ cm and a period $T = 140 \pm 2$ s). Spirals were created at the center of the medium to avoid boundary influence [11]. Note that, due to the generation method, both spirals do not have exactly mirror symmetry at the beginning of the experiment. We can define the asymmetry degree as the difference between the distances of spirals to the shock line divided by the least of them. With this definition, we can say that two spirals are almost symmetrical if their asymmetry degree is less than 10%. The experiments were followed with a CCD camera connected to a Silicon Graphics workstation where images were digitized and spiral tip positions were automatically measured and stored every 3 sec. These recorded tip positions allow us to determine the positions of the center of the cores of both spirals and also the angle between cores. From their temporal evolution, the mean drift velocity of each spiral can be determined. We will separate mean drift velocity into radial V_r (in the direction of the line connecting cores) and tangential V_t (perpendicular to radial line) components.

Although we have mentioned the relative distance between cores in the description of interaction ranges, this parameter is not enough to explain satisfactorily why a spiral dominates the other one. The asymmetry of the initial pattern seems to be the determining factor. The relative distance between cores does not characterize perfectly two unlike interacting spirals. So, for a given relative distance, spirals may be symmetrical or asymmetrical (the distance does not need to be uniformly distributed between them). We have found that the distance from each core to the shock line is a good parameter to describe the initial symmetry and, thus, the observed behavior. Figure 2 shows the temporal evolution of the distance to the shock line (we will call this distance DsD for the dominant spiral and Dsd for the dominated one) for the spirals shown in Fig. 1. The spiral placed farther from the shock line is the one which dominates, and the other, initially closer, is dominated. Spirals have a slight initial asymmetry, which makes their distance to the shock zone to be different. The mean frequency of the spiral farther from the shock line (and thus dominant) is measured to be higher, which is related to the fact that this spiral develops more wavelengths than the dominated one. During the first hour, $\langle R_d \rangle$ hardly varies [see also Fig. 1(d)], whereas the distance of each spiral to the shock line changes. The dominant spiral goes away from the shock zone, and thus its distance to the shock line increases in time. For the dominated spiral, this distance is observed to decrease until a value of around 0.4λ, and then to remain constant around that value. This spiral is only a bare core and, therefore, its distance to the shock zone can be reduced no more.

In our experiments, there always appears a drift movement superimposed to the rotation around the core (movement of a single spiral in an infinite medium). We will show that asymmetry is a determining factor in the way spirals drift through the medium. If each spiral is far enough (several wavelengths) from the shock zone and they are nearly symmetrical, a particular correlation between mean drift velocity components is observed. In Figs. 3(a) and 3(b), these radial and tangential components are represented for the case of two unlike spirals

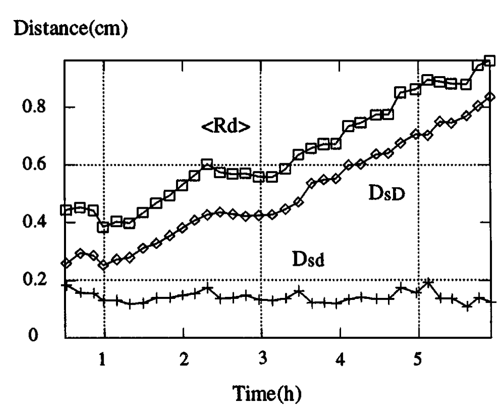

FIG. 2. Temporal evolution of the distance to the shock line (DsD for the dominant spiral and Dsd for the dominated one) for spirals shown in Fig. 1 ($\lambda \approx 0.3$ cm). $\langle R_d \rangle$ represents the mean relative distance between spiral tips. Data points have been taken approximately every 10 minutes (every four collisions).

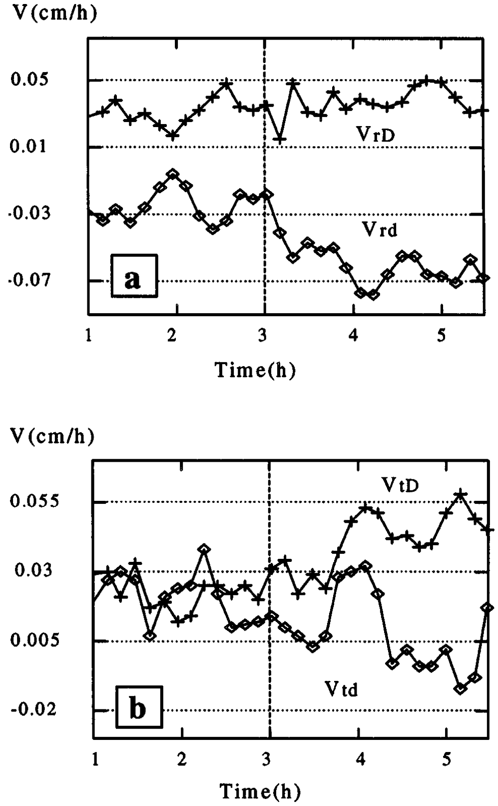

FIG. 3. Temporal evolution of radial (a) and tangential (b) components of the mean drift velocity of two interacting spirals. At first they are almost symmetrical and the mean distance between their tips is of around 4λ ($\lambda \approx 0.3$ cm). During the experiment spiral on the left dominates spiral on the right but it is not able to reduce it to its bare core (at least during the allowed experimental time).

initially placed at a distance of $\langle R_d(t = 0)\rangle \approx 4\lambda$ (both spirals are nearly symmetrical and each one is separated two wavelengths from the shock line). It becomes apparent that radial velocities are equal in modulus and opposite in direction ($V_{rD} \approx -V_{rd}$, with D denoting the dominant spiral and d the dominated one), while tangential ones are identical both in modulus and in direction ($V_{tD} \approx V_{td}$). As time goes by, this correlation is observed to change at the same time that one of the spirals dominates. After 3 hours from the beginning of the experiment, marked by the dashed line in Fig. 3, the reported correlation stops being valid.

Different studies of spiral interaction in the framework of the Ginzburg-Landau equation [17–20] have found that two symmetrical spirals with different chirality drift in a way similar to that observed in Fig. 3. Theoretical approaches [17–19] have obtained opposite radial and equal tangential velocities, providing spirals are too far apart (therefore fulfilling the condition that the perturbation in spiral amplitude induced by interaction is small). So, the predicted correlation for radial and tangential components of drift velocities of two symmetrical spirals is

$$V_{t1} = V_{t2}, \qquad V_{r1} = -V_{r2} \tag{1}$$

(note that we do not talk of dominant D and dominated d spiral because spirals are symmetrical). Numerical simulations of interacting symmetrical opposite spirals both in the GL equation [20] and in the FitzHugh-Nagumo system [21] have reported this same behavior for not so long distances between spirals (in [21] symmetrical spirals with this behavior are initially separated a distance $1.5d$, with d being the diameter of the core). In fact, the reported correlation can be predicted by symmetry arguments [17]. In our experiments, though at first we observe that correlation, it is lost as asymmetry grows.

In fact, the correlation between drift velocity components is found to vary with the asymmetry between spirals. So, a different correlation between drift velocity components is obtained when one of the spirals is totally reduced to its core. Tangential and radial components of drift velocity of spirals in the experiment shown in Fig. 1 [$\langle R_d(t = 0)\rangle \approx 1.6\lambda$] are plotted in Fig. 4(a). The radial

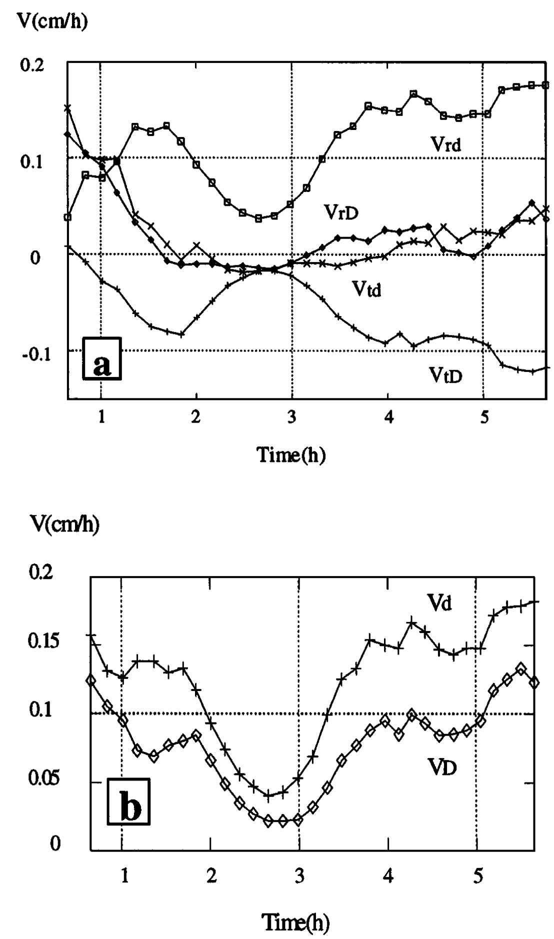

FIG. 4. Temporal evolution (a) of tangential and radial components of the mean drift velocity of spirals shown in Fig. 1 ($\lambda \approx 0.3$ cm). The modulus of this mean drift velocity is shown in (b). Note that there appears a minimum about 3 hours from the start, which coincides with the moment when spirals are in phase.

drift velocity of the dominant spiral is found to exhibit a behavior similar to the tangential component of the dominated one ($V_{rD} \approx V_{td}$). It can also be seen that the sum of the tangential component of the dominant spiral and the radial component of the dominated one is a constant ($V_{tD} + V_{rd} \approx$ const). It is remarkable that this reported form of correlation stands when one of them has dominated and reduced the other one to its bare core—note in the figure that this correlation is clearly valid from 1 hour on, when spirals are already quite asymmetrical. The modulus of the drift velocity of both spirals is represented in Fig. 4(b). The functional evolution of both velocities is quite similar, but the velocity of the dominated spiral is always higher. Therefore, in this case, the drift movement of the dominated spiral is faster than that of the dominant one.

In summary, we have observed the existence of different correlations between the drift velocity components of both spirals depending on the pattern symmetry. Thus, when spirals are slightly asymmetrical and their distance to the shock line is longer than several wavelengths, they behave as predicted for a symmetrical state in the literature [17–21]. In this situation, the existence of wave fronts between both spirals and the shock line "*screens*" the interaction, and the symmetrical correlation stands. However, when the asymmetry is big enough (Dsd is short enough—close to a single wavelength—and quite different from DsD) the theoretical prediction is valid no more and the way spirals drift changes in time. Finally, when one of the spirals is completely dominated and reduced to its bare core, we find a new stationary correlation coupling radial and tangential velocities of both spirals. Now, the dominated spiral is reduced to its core and, therefore, it has no wave fronts that screen the interaction. Consequently, it moves faster than the dominant one [see Fig. 4(b)], which has developed some wavelengths.

Finally, although it could be thought that the observed behavior is due to inhomogeneities present in the medium, we have shown that spirals drift through the medium, and thus a local perturbation like an inhomogeneity would not influence spiral behavior during a long time. At most, it may induce an asymmetry and, thus, domination, but the perturbation would be a short-term one, because the spiral goes away from the inhomogeneity in its drift. We believe that interaction between spirals is an effect similar to interaction between a spiral and a boundary. The presence of a spiral, like the presence of a boundary, causes the medium to be anisotropic and, therefore, influences the spiral dynamics (in [11] a geometrical-kinematical model is presented that suggests a mechanism to explain how the anisotropy induced by the presence of the boundary gives rise to the drift of a spiral). In fact, translation symmetry is lost, and each spiral acts as if it encountered an impenetrable boundary in the shock zone, but a boundary changing from shock to shock. The typical order of magnitude of both spiral-spiral interaction and boundary-spiral interaction is similar (drift induced velocities are of the order of 1% of linear rotation velocity), in accordance with what would be expected if they had the same origin.

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