

Experimental Determination of the Dispersion Relation for Spiral Waves

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We have experimentally measured the dispersion relation of spiral waves in the Belousov-Zhabotinsky reaction in an open spatial reactor, for different concentrations. Appropriate rescaling permits the collapse of all data onto a single line. From this, we infer an analytic form for the dispersion relation, which only depends on two physical quantities. In particular, this relation implies that the ratio between the front velocity of the selected spiral and the isolated front velocity is fixed and equal to 0.62, in agreement with previous results. [S0031-9007(96)00797-1]

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Reaction-diffusion systems have become a fundamental part of the study of nonequilibrium phenomena since Turing first suggested that they were sufficient to account for biological pattern formation [1]. The spiral wave is a stable rotating pattern observed in many reaction-diffusion systems, particularly in excitable media [2–5]. Such dissipative media, which include nerve axons [6] and cardiac muscle [7,8], can sustain a propagating constant amplitude wave front, when locally excited beyond a threshold. The well-known Belousov-Zhabotinsky (BZ) chemical reaction [9] can provide a laboratory realization of an excitable medium.

It is relatively easy to prepare a small layer of BZ solution in a Petri dish, to induce a wave front, and to break it. After a few minutes, two spirals of opposite chirality develop, which usually rotate at a constant rate. The geometrical purity of these patterns is fascinating and much experimental as well as theoretical work has been devoted to understanding their formation, characteristics, and properties [10–12].

Despite these efforts, a number of questions remain open. Among them, knowledge of the dispersion properties of this wave sustaining medium is still imprecise and essentially relies on numerical or analytical studies; few conclusive experimental results are available [13]. Nevertheless, the dispersion relation plays an essential role in the selection mechanism by which spirals in a given medium have a unique pitch and period.

There are two principal reasons for the rarity of experimental results. First, most experiments have been conducted in closed systems, in which true stationary states cannot be observed, and fast rotation rates are not obtainable. This represents a serious drawback for quantitative measurements. The other reason comes from the difficulty of imposing a different period and pitch on a spiral. We present here the measurement of the dispersion relation for spiral waves made in the BZ reaction in an open spatial reactor. The experimental key point relies on the local sensitivity of the ferroin-catalyzed reaction to red laser light [14]: locally applying this light slows

down the reaction. Hence when the beam is focused on the core region of the spiral, its pitch increases and it spins more slowly. This technique has already been used in a slightly different version of the BZ reaction [15], but with the limitations of a closed reactor.

Our experimental setup has already been described [16]. It is made of two continuously fed well-stirred tank reactors (CSTR A and CSTR B) separated by a thin porous glass disk, 0.4 mm thick and 25.4 mm in diameter (Vycor glass, Corning). Sulfuric acid and sodium bromate are fed into both CSTRs, whereas malonic acid (MA) is present only in CSTR A and ferroin only in CSTR B. A small amount of sodium dodecyl sulfate (SDS) and sodium bromide fed into CSTR A prevents large bubble formation and bulk oscillation of the reactor. Hence pattern forming waves occur only inside the porous glass between the two chambers. The system is thus fairly two dimensional and of pure reaction-diffusion nature. For our measurements, the concentrations used are $[\text{NaBrO}_3]_0 = 0.40M$, $[\text{MA}]_0^A = 0.40M$, $[\text{NaBr}]_0^A = 30 \text{ mM}$, $[\text{SDS}]_0^A = 0.03 \text{ mM}$, and $[\text{ferroin}]_0^B = 1.0 \text{ mM}$; $[\text{H}_2\text{SO}_4]_0$ took four different values: 0.2M, 0.3M, 0.4M, and 0.5M (scans I to IV, respectively). An additional series of measurements (scan V) was performed for $[\text{H}_2\text{SO}_4]_0 = 0.40M$, $[\text{NaBrO}_3]_0 = 0.15M$, and $[\text{MA}]_0^A = 0.20M$; the other concentrations were as before. The superscript, when present, designates the CSTR into which the chemical is introduced. These concentrations have been chosen so that a simply rotating spiral is obtained, far from any bifurcation or instability [17]. The residence time of the chemicals in the CSTRs is 15 mn and the temperature is regulated at $23 \pm 0.1 \text{ }^\circ\text{C}$.

The procedure to create a single spiral in the reactor is the following. When the chambers are first filled, the edges of the porous glass start to emit waves. The attenuated light beam of a red laser (either a diode laser, 3 mW, $\lambda = 670 \text{ nm}$, or a helium-neon laser, 1 mW, $\lambda = 633 \text{ nm}$) is focused onto the porous glass to a spot size of about 100 μm . This locally inhibits the BZ reaction, and breaks the wave front, creating two topological defects that evolve



FIG. 1. Snapshot of the evolution of the spiral, 3 periods after the laser has been turned on. Though hardly visible, the laser spot is placed at the spiral core. The circular edge of the cell is visible, which has a diameter of 20.25 mm. Concentrations $[\text{H}_2\text{SO}_4]_0 = 0.3M$, $[\text{NaBrO}_3]_0 = 0.40M$, $[\text{MA}]_0^A = 0.40M$, $[\text{NaBr}]_0^A = 30 \text{ mM}$, $[\text{SDS}]_0^A = 0.03 \text{ mM}$, and $[\text{ferroin}]_0^B = 1.0 \text{ mM}$.

into two spirals of opposite chirality. We then use the laser to move one of the spirals off the edge, and center the remaining one.

Once the spiral pitch is uniform over the whole cell, i.e., the spiral has reached its *selected state*, we wait for at least 50 spiral periods before beginning to measure the dispersion curve. Since the inhibitory effect of the laser beam depends on the impinging intensity, we use several filters in different combinations to produce a range of intensities [18]. When the laser beam is focused at the spiral core, this local inhibition induces an increase of the period and pitch near the tip. Figure 1 illustrates the transient shape which occurs after the laser beam is turned on. After a few periods, the entire spiral has adjusted to its new pitch. Once it has reached a steady state, the period is measured with a stop watch and the pitch with an image processing program (for a description, see [19]). The precision for all measurements is always better than 5%. When the laser beam is removed, the medium recovers and the spiral returns to its selected state. Figure 2 shows a spiral at its selected pitch and period [Fig. 2(a)], and its asymptotic state when submitted to three different laser intensities [Figs. 2(b)–2(d)].

For each scan, the pitch p and period T constitute the raw data. Since curvature effects are negligible beyond a few turns from the spiral tip [20], we can write the normal wave velocity as $c = p/T$. In Fig. 3, we plot c vs T for scan V. The velocity tends to saturate for large T , as expected in the case of excitable media, where successive wave fronts become independent of each other for large enough spacing. The solid curve arises from the analysis of our data, and is explained below.

In order to compare spirals obtained under different experimental conditions, we consider the dimensionless number $M \equiv p^2/DT$, where D is the diffusion constant of the

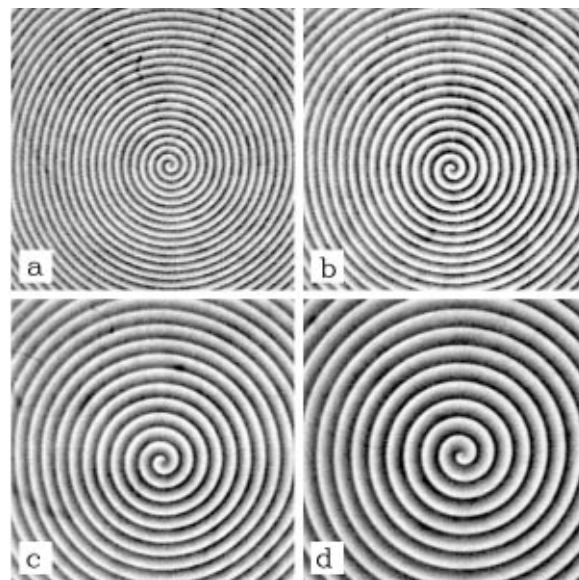


FIG. 2. Example of the influence of a laser beam on a spiral, as impinging intensity is increased. Measured (p, T) : (a) selected spiral ($344 \mu\text{m}$, 5.83 s); laser on: (b) ($450 \mu\text{m}$, 6.50 s); (c) ($575 \mu\text{m}$, 7.77 s); (d) ($720 \mu\text{m}$, 9.34 s). $[\text{H}_2\text{SO}_4]_0 = 0.5M$; all other parameters as in Fig. 1. Images are 11.5 mm across.

medium. We use $D = 2.0 \times 10^{-5} \text{ cm}^2/\text{s}$, as measured for the BZ components [21]. This number M is the ratio of the two relevant time scales in the problem, the diffusion time across the spiral pitch (p^2/D) to the rotation period T . Both M and its inverse have been measured in the BZ reaction [22] and in numerical simulations [23–25]; it was

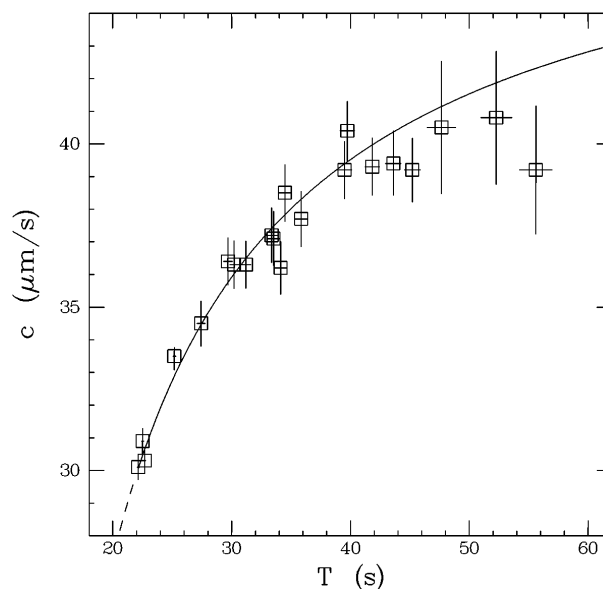


FIG. 3. Wave speed c as a function of wave period T ; the solid line corresponds to Eq. (2). The point for the selected spiral is at the lower left, which has the values $c_s = 30.4 \mu\text{m/s}$, and $T_s = 22.1 \text{ s}$. Concentrations $[\text{H}_2\text{SO}_4]_0 = 0.4M$, $[\text{NaBrO}_3]_0 = 0.15M$, $[\text{MA}]_0^A = 0.20M$; other concentrations as in Fig. 1.

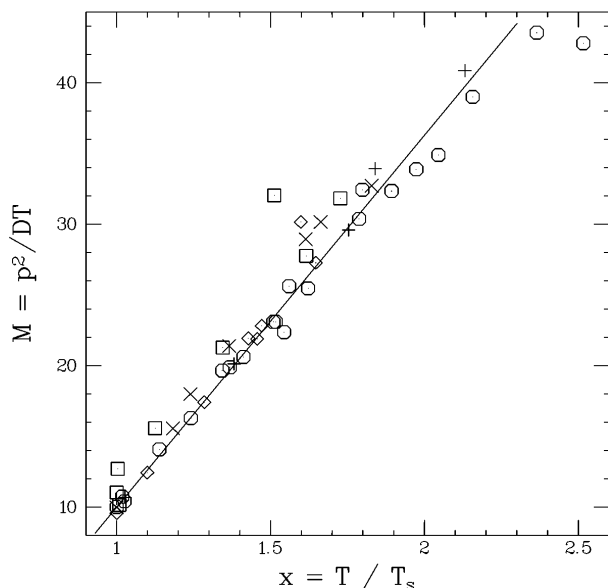


FIG. 4. Dimensionless numbers $M \equiv p^2/DT$ vs $x \equiv T/T_s$. Symbols correspond to the different scans: +, scan I; \times , scan II; \diamond , scan III; \square , scan IV; \circ , scan V. The solid line corresponds to Eq. (1).

first introduced in models of spiral formation [26,27]. In Fig. 4 we plot M for all five scans vs the normalized period $x = T/T_s$, where T_s is the selected rotation period. The five curves collapse onto a single straight line. Note that T_s is a characteristic of the medium and changes for each scan; it ranges from 18.5 s (scan I) to 5.8 s (scan IV), and is equal to 22.1 s for scan V. Hence Fig. 4 is a *universal dimensionless plot* that describes all of our data. The selection is given by $x = 1.0$, at which $M \approx 10$. Our results show that, as the spiral is slowed down, its internal diffusion constant (p^2/T) is increased: this curve represents an effective diffusion enhancement.

A linear fit to the data in Fig. 4 leads to

$$M = \beta x - \alpha, \quad \text{with } \alpha = 16.3, \quad \beta = 26.3. \quad (1)$$

From Eq. (1), we can derive an expression for the dispersion relation,

$$c(T) = c_\infty(1 - T_0/T)^{1/2}, \quad (2)$$

with $c_\infty = (\beta D/T_s)^{1/2}$, $T_0 = T_s \alpha/\beta$.

This functional form introduces two dimensional quantities: the asymptotic velocity c_∞ which is interpreted as the velocity of an isolated single wave in the medium and the time scale T_0 for which the extrapolated wave velocity would vanish. The physical meaning of T_0 is unclear since it is known that waves become unstable below a finite velocity [13]. We speculate that T_0 corresponds to some chemical time scale of the system. For the data in Fig. 3 (scan V), we get $c_\infty = 48.8 \mu\text{m/s}$ and $T_0 = 13.6$ s. It should be noted that c converges slowly to c_∞ as T increases: it is within 5% of its asymptotic

value for $x \approx 6$, and within 1% for $x \approx 31$, whereas our data only go up to $x = 2.5$.

Since α and β are two fixed numbers, we obtain a fixed ratio between the velocity at the selection c_s and the asymptotic velocity, namely,

$$c_s/c_\infty = (1 - \alpha/\beta)^{1/2} = 0.62. \quad (3)$$

Previous experimental measurements for c_s/c_∞ give 0.61 [21] and 0.67 [23]. Reference [15] provides an upper bound of 0.67. A value of about 0.6 is also found for an approximation of this ratio in a compilation of experimental data [13]. The ratio between T_0 and T_s is also fixed and equal to $\alpha/\beta = 0.62$. We have no explanation for the size of these ratios, or the fact that they are equal.

In the BZ reaction, several previous measurements of the dispersion relation have been made [13,15,21,23,28]; it was also measured for crescent-shaped waves in a pinwheel reactor [29]. With the exception of this last experiment, all studies were made in closed chemical reactors. In most cases only a single set of concentrations was used; one study compared four different concentrations, but attempts at rescaling did not lead to a single curve [13]. The only unambiguous common feature of these experimental dispersion curves is a shape qualitatively similar to Fig. 3.

Two techniques have been used to theoretically determine the dispersion relation: singular perturbation theory [27,30] and direct numerical simulation of the Oregonator, a two variable dynamical system arising from a chemical model of the BZ reaction [23,24]. The latter studies should permit a quantitative connection to the chemistry. They suggest a possible rescaling of all dispersion curves, but two parameters cannot be precisely related to the experiment, and the comparison fails. The analytical approaches consider two reaction-diffusion partial differential equations in the limiting case where one variable evolves much faster than the other. The dispersion relation is obtained by constraining the two fronts forming the wave to propagate with the same velocity. Unfortunately this calculation does not lead to a formula, and the dispersion curve must be obtained by numerical solution. In addition, the results are usually presented in terms of internal theoretical quantities which are not experimentally accessible. Hence no direct comparison with these theories can be made. A single analytical form for the dispersion relation has been suggested in [31], but without any theoretical justification. Our results provide an experimentally motivated analytical form in terms of measurable quantities. Although Eq. (3) is not derived from a microscopic analysis of the chemical processes, we believe that they are hidden in the only two parameters, namely, the diffusion constant and the spiral period at selection.

As an additional result of our analysis, we see from Fig. 4 that the selected values of the pitch and period always occur at a constant value $M_* \approx 10$. Published values of this number coming either from experiments in closed systems or from simulations vary between 17 and 900 [22,24], but theories first developed for crystal growth

predict that it should be constant, and approximately equal to 20 [26,27]. This arises from the geometrical constraint that a spiral is a stationary rotating object with a constant pitch. A systematic study of the spiral characteristics over an extensive range of the parameter space also showed that M is constant and equal to M_* [17]. Hence our result agrees with the theoretical prediction. The numerical discrepancy in the amplitude of M_* might simply be due to imprecision in D .

We have experimentally studied a simple dynamical pattern, a rotating spiral wave, and used it to measure the dispersion relation for the BZ reaction. We found that the dimensionless ratio $M = p^2/DT$ varies linearly with the reduced period T/T_s , independent of concentration. This has led us to an analytic form for the velocity $c(T)$, Eq. (2). It implies a dimensionless ratio, Eq. (3), which is in agreement with previous results. Theoretically, it now remains to derive this equation from general considerations for reaction-diffusion media. Experimentally, further studies are needed to extend the validity of the dispersion relation: to check its slow approach to saturation, longer periods must be obtained, either with a stronger inhibition at the spiral core, or by another technique. Future quantitative measurements will tell to what extent our dispersion relation is universal; could one observe a spiral which does not obey this relation? Also, we have only examined simply rotating spirals; it is not clear how meandering spirals will fit into this picture. However, since the selected spiral period T_s and the diffusion constant D are the only free parameters in our description, from which all other quantities can be derived, they somehow contain all the information of the reaction-diffusion medium. This provides a strong impetus towards some future comprehensive theory for spiral dynamics, and perhaps for general pattern formation in reaction-diffusion systems.

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Note added.—There are also other functional forms compatible with our scaled data, which still give a ratio $c_s/c_\infty \approx 0.62$. In particular, the function $c(T) = c_\infty(1 - \exp[-T/T_s])$ provides a good fit, and gives $c_s/c_\infty = 1 - 1/e \approx 0.635$. It does not, however, imply a minimum period below which propagation is impossible. We thank V. Gáspár for pointing this out to us.

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