

## Wave propagation in an excitable medium along a line of a velocity jump

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The propagation of an excitation wave in a distributed medium along a stripe with increased velocity is shown to result in the formation of a stationary V-shaped wave structure. The propagation velocity of this structure depends on the width of the stripe due to effects of wave curvature. We observed this phenomenon in a light-sensitive version of the Belousov-Zhabotinsky system under nonhomogeneous illumination. Equations are derived that describe quantitatively the observed wave structures and applied to estimate the diffusion coefficient of the propagator species.

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A wide class of self-organization processes in complex physicochemical systems is closely related to wave propagation in so-called excitable media [1–3]. A distributed excitable medium [e.g., the chemical Belousov-Zhabotinsky (BZ) reaction [1] or heart muscle [2]] consists of locally coupled active elements which can form a pulse in response to an external signal. Such pulses travel through the medium as autowaves. Their properties differ significantly from acoustic or electromagnetic waves in conservative systems. For instance, they annihilate under collisions, are not reflected from boundaries, and their velocity depends on the local wave-front curvature.

We report the observation of a stationary-wave pattern propagating along a thin stripe with higher velocity placed in a homogeneous medium with lower velocity. The wave front in a region surrounding the stripe is tilted with respect to the propagation direction but it is flat, whereas the front inside the stripe is strongly curved. Hence such an experiment produces an effective and simple means to study the curvature-speed relationship. In particular, we have measured the dependence of the propagation velocity of the V-shaped structure on the width of the stripe and then elaborated a mathematical description of the experimental results using the kinematical theory of autowave patterns [4,5]. This leads to an estimate of the important phenomenological parameter  $D$ , which is the slope of the linear dependence of autowave velocity on local curvature.

Experiments were performed by using a light-sensitive version of the BZ reaction. In this system the reduced state of the catalyst rutheniumbipyridyl [Ru(II)] promotes the autocatalytic production of the crucial propagator species  $\text{HBrO}_2$ . Once this complex is photochemically excited to  $\text{Ru(II)}^*$  it catalyzes the production of the inhibitor  $\text{Br}^-$  [6,7]. Using this mechanism it is possible to control the local excitability of the system by the intensity of the applied illumination [8–11]. In particular, the illumination decreases the velocity of a wave.

In order to avoid hydrodynamic perturbations, a silica gel (thickness 0.6 mm, diameter 7 cm) was used as a matrix of the reactive solution in which the catalyst  $\text{Ru}(\text{bpy})_3^{2+}$  (4 mM) was immobilized [12]. Disregarding

the bromination of malonic acid, the initial reactant concentrations in the gel are calculated as follows: 0.09M NaBr, 0.2M  $\text{NaBrO}_3$ , 0.17M malonic acid, and 0.47M  $\text{H}_2\text{SO}_4$ . The temperature was kept fixed at  $(24 \pm 1)^\circ\text{C}$ . In this system waves propagate for approximately 3 h. To avoid drift effects due to slow aging of the solution the measuring times were kept as short as possible.

The observation light, which also controlled excitability, was emitted from a high-pressure mercury lamp (150 W). Two-dimensional transmission through the reactive layer was recorded by a charge-coupled-device camera (Hamamatsu C3077) at 490 nm and stored on a time-lapse video recorder. Single frames of the resulting movies were digitized by an image-acquisition card (Data-Translation DT-2851) and finally analyzed on a personal computer.

We induced in the experiments almost planar wave trains of constant period ( $T = 35 \pm 1$  s) propagating in an illuminated medium. By using black stripes, which blocked the observation light, we created a region with a higher excitability and, correspondingly, with faster wave propagation. The stripes were oriented perpendicularly to the wave fronts. Their length was approximately 4 cm with half widths (half the value of the width of the stripe)  $W$  of 0.1–2.1 mm. Due to the different propagation velocity inside and outside the stripe the planar waves transformed to V-shaped ones in the course of time (see Fig. 1).

Characteristic properties of stationary-wave propagation in this inhomogeneous medium were measured for different half widths  $W$  of the dark stripes. Five or six stripes with increasing  $W$  were used in succession during one experiment. Each measurement was performed after a waiting period of 150 s, during which the pattern achieved a new steady geometry. Figure 2 shows characteristic experimental snapshots taken immediately after removing the dark stripes. The leading, highly curved portion of the front is located inside the well excitable region, whereas the fronts in the unperturbed illuminated surroundings are tilted with respect to the initial front orientation. The angle  $\beta$  between the tilted front and the planar wave increases with increasing values of half width  $W$ . Figure 3 describes the dependence of the wave

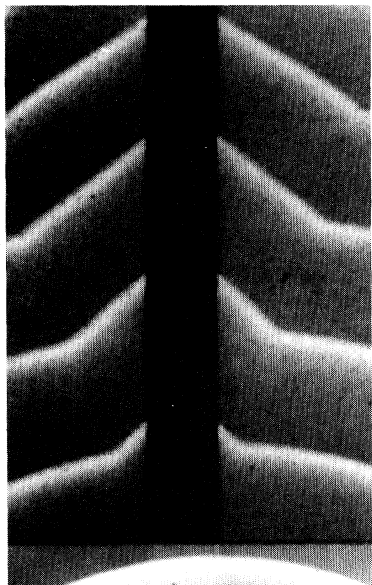


FIG. 1. Waves of excitation in a light-sensitive BZ system propagating along a dark stripe (half width 1.0 mm) with higher velocity. Initially planar waves become V-shaped and achieve a new steady geometry.

velocity  $U$  along the stripe boundary on half width  $W$  using the data obtained from two typical experiments. The value of  $U$  for  $W=0$  is the normal velocity of planar fronts in the homogenous, illuminated medium ( $U \approx 82 \mu\text{m/s}$ ). The velocity  $U$  increases rapidly with  $W$  and saturates at approximately  $97 \mu\text{m/s}$ . The velocities are derived during a 21-s interval from waves that have passed already 1.5 cm along the dark stripe. Careful analysis

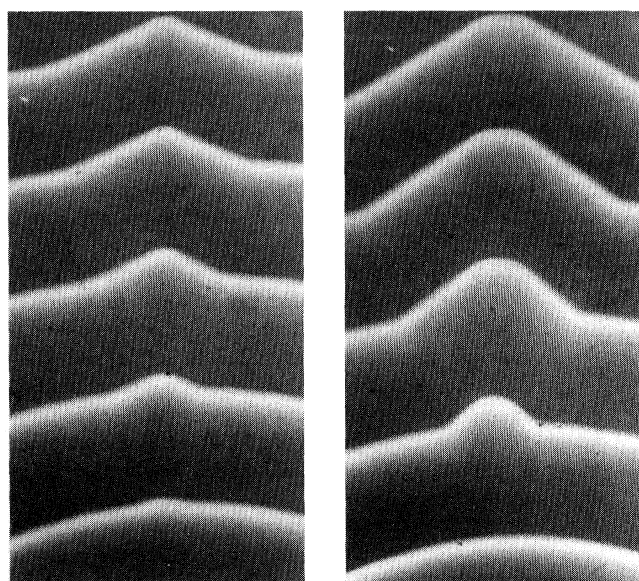


FIG. 2. Shapes of autowaves observed immediately after removing the dark stripes of different half width. Half width  $W$  and observed angle  $\beta_W$ : (a)  $W=0.28$  mm,  $\beta_W = -26.4^\circ$ ; (b)  $W=0.83$  mm,  $\beta_W = -35.7^\circ$ .

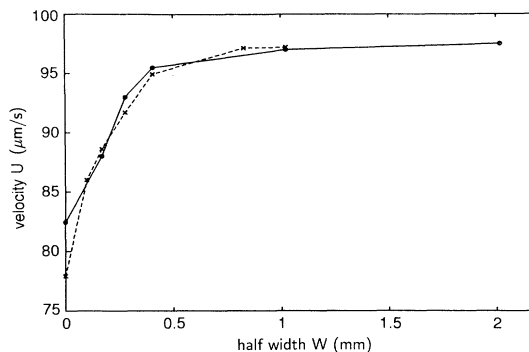


FIG. 3. Dependence of propagation velocity  $U$  on stripe half width  $W$  obtained in two different experiments.

confirms that the velocity  $U$  and the angle  $\beta$  at this distance have increased to steady values.

The pattern in Fig. 1 is similar to shock-wave structures, for instance, created by sound propagation along a wire surrounded by a liquid [13]. For such an acoustic system the sound velocities  $V_1$  in the wire and  $V_2$  in the liquid are constant, with  $V_1 > V_2$ . In the liquid one can observe flat acoustic waves tilted with respect to the wire by an angle  $\beta$ , which satisfies the equation  $\cos\beta = V_2/V_1$ . We emphasize, however, that in our case the propagation velocity along the high-velocity stripe is not constant, but depends on the width of the stripe.

The theoretical explanation of the observed phenomena is based on the kinematical description of autowaves [4,5]. Let us consider the autowave front propagating through a long excitable channel ( $-L \leq x \leq L$ ) with impermeable boundaries and with a stripe of high velocity placed on the symmetry axis ( $-W \leq x \leq W$ ) (see Fig. 4). The normal velocity  $V$  of each point of the front depends on the local curvature  $K$ :

$$V = V_0 + DK. \quad (1)$$

Here  $V_0$  is the velocity of the planar autowave. For

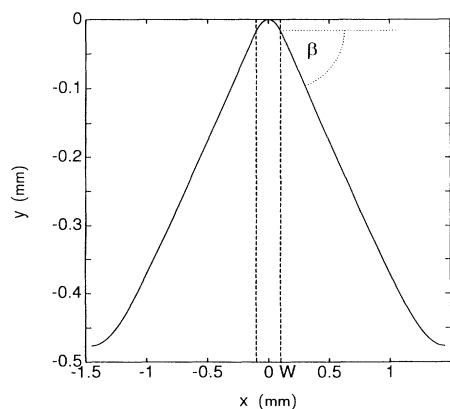


FIG. 4. Stationary shape of a wave front computed from the analytical solution (6) and (7) of Eq. (4). Parameter values:  $V_d = 97.9 \mu\text{m/s}$ ,  $V_l = 77.9 \mu\text{m/s}$ ,  $U = 83.9 \mu\text{m/s}$ ,  $\beta_W = 17.9^\circ$ ,  $L = 1.44$  mm,  $W = 0.1$  mm,  $D = 4.5 \times 10^{-5} \text{ cm}^2/\text{s}$ .

the BZ system, coefficient  $D$  in (1) is of the same order as the diffusion coefficient of the propagator species  $\text{HBrO}_2$  [14,15], which is about  $2 \times 10^{-5} \text{ cm}^2/\text{s}$ . In particular, for the medium with fixed catalyst, the value of  $D$  should be larger [4,5]. The value  $V_0$  differs for different parts of the investigated medium:  $V_0 = V_d$  in the dark stripe ( $-W \leq x \leq W$ ) and  $V_0 = V_l$  in the illuminated region ( $|x| \leq L$  and  $|x| > W$ ). Due to this difference the initially flat wave front should be curved with time. Let us assume that a stationary shape of the front is achieved after a relaxation process and  $U$  is the propagation velocity of the stationary structure as a whole along the  $y$  axis (see Fig. 4).

One can express the normal velocity  $V$  as

$$V = U \cos \beta, \quad (2)$$

where  $\beta$  is the angle between the  $x$ -axis and the tangent to the front.

Inserting (2) into (1) we obtain

$$DK = U \cos \beta - V_0, \quad (3)$$

Since  $K = d\beta/ds$ , where  $s$  is the arclength, we finally have

$$\frac{d\beta}{ds} = (U \cos \beta - V_0)/D. \quad (4)$$

Equation (4) has an exact analytical solution, but the form of this solution differs from the cases  $U < V_0$  and  $U > V_0$ . Hence we need to describe the parts of the front inside and outside the dark stripe in different ways. For this it is convenient to rewrite (4) using Cartesian coordinates:

$$\begin{aligned} \frac{d\beta}{dx} &= \frac{U \cos \beta - V_0}{D \cos \beta}, \\ \frac{d\beta}{dy} &= \frac{U \cos \beta - V_0}{D \sin \beta}. \end{aligned} \quad (5)$$

Due to the symmetry of the pattern it is sufficient to find the solution of (5) only for the right half of the medium.

For the dark stripe we have  $V_0 = V_d > U$ . For this case we can integrate (5) with initial conditions  $x=0$  and  $y=0$  for  $\beta=0$  and present the shape of the front in parametric form:

$$\frac{x}{D} = \frac{\beta}{U} - \frac{2V_d}{U\sqrt{V_d^2 - U^2}} \arctan \frac{(V_d + U)\tan(\beta/2)}{\sqrt{V_d^2 - U^2}}, \quad (6a)$$

$$\frac{y}{D} = \frac{1}{U} \ln[(U - V_d)/(U \cos \beta - V_d)]. \quad (6b)$$

Here,  $x$  and  $y$  are the Cartesian coordinates of the front within the dark stripe (i.e., for  $x < W$ ).

For a medium surrounding the dark stripe ( $W < x < L$ ) we have  $V_0 = V_l < U$ . In this region the solution of (5) is

$$\frac{x - W}{D} = \frac{\beta - \beta_W}{U} + \frac{V_l}{U\sqrt{U^2 - V_l^2}} \ln \frac{\chi(\beta)}{\chi(\beta_W)}, \quad (7a)$$

$$\frac{y - y_W}{D} = \frac{1}{U} \ln \frac{U \cos \beta_W - V_l}{U \cos \beta - V_l}, \quad (7b)$$

$$\chi(\beta) = \frac{\sqrt{U^2 - V_l^2} + (U + V_l)\tan(\beta/2)}{\sqrt{U^2 - V_l^2} - (U + V_l)\tan(\beta/2)}, \quad (7c)$$

where  $\beta_W$  and  $y_W$  are the values of  $\beta$  and  $y$  at the boundary of the dark stripe, i.e., for  $x = W$ .

The front should be orthogonal to an impermeable boundary. Hence, the angle  $\beta=0$  for  $x=L$ . Putting this boundary condition into Eq. (7a) we get

$$\frac{L - W}{D} = \frac{V_l}{U\sqrt{U^2 - V_l^2}} \ln \frac{1}{\chi(\beta_W)} - \frac{\beta_W}{U}. \quad (8)$$

The shape of the stationary front computed from the system (6)–(8) is shown in Fig. 4. This characteristic V-shaped front is stationary and similar to the wave structures observed experimentally (Figs. 1 and 2). In our experiments the size of the medium is much larger than the width of the stripe. The corresponding calculations for the case  $L \gg W$  show that the shape of the front in the region surrounding the dark stripe is close to a straight line and curved only near the boundary of the medium (see Fig. 4). The angle  $\beta_W$  depends on  $L$  and the function  $\chi(\beta_W)$  decreases with  $L$ . Moreover, for the case of an unbounded medium ( $L = \infty$ ) the function  $\chi(\beta_W)$  vanishes and the curve defined by (7) degenerates into the straight line (similar to the case of shock waves):

$$\beta = \beta_W^* = -\arccos(\theta_l/U). \quad (9)$$

Note that the function  $\beta_W(L)$  saturates very rapidly with  $L$ . For instance, the angle  $\beta_W$  calculated for  $L = 1.44 \text{ mm}$  (see Fig. 4) differs from the asymptotic value (9) only by 0.01%. Hence we can use this asymptotic value of  $\beta_W$  to estimate the dependence of the propagation velocity  $U$  on the width of the dark stripe which was observed experimentally.

Since  $\beta$  is a continuous function of the coordinate  $x$ , we substitute  $x = W$  and  $\beta_W = \beta_W^*$  from (9) into (6a). Then we get the dependence of the propagation velocity  $U$  on the half width  $W$  in the following form:

$$W = \frac{D}{V_d} \left\{ \frac{\beta_W}{u} - \frac{2}{u\sqrt{1-u^2}} \times \arctan \left[ \frac{1+u}{\sqrt{1-u^2}} \tan(\beta_W/2) \right] \right\}, \quad (10)$$

where  $u = U/V_d$  and  $\beta_W = -\arccos(V_l/U)$ .

According to (10) the velocity  $U$  of the stationary autowave structure is an increasing function of half width  $W$ . The single free parameter in (10) is the unknown value of coefficient  $D$ . By fitting this parameter to the experimental data we find  $D = 4.5 \times 10^{-5} \text{ cm}^2/\text{s}$ . The corresponding dependence of  $U$  on  $W$  is plotted in Fig. 5. For this value of  $D$  the theoretical results are in a good quantitative agreement with the experimental data presented in Fig. 3. Furthermore, in Fig. 6 we used the same value of the coefficient  $D$  to plot the front shape.

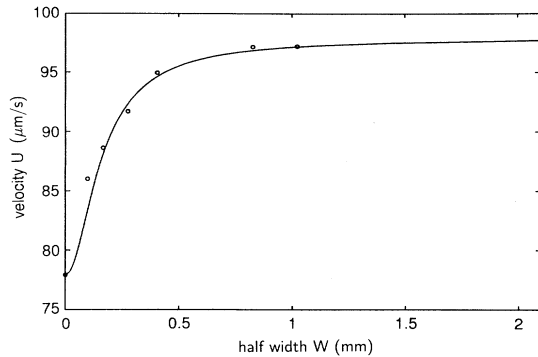


FIG. 5. Dependence of propagation velocity  $U$  on stripe half width  $W$  fitted by Eq. (10) (solid line) to one set of the experimental data presented in Fig. 3.

The results shown in Figs. 5 and 6 demonstrate that the kinematical theory provides a remarkably good description of the observed phenomenon. Moreover, the presented procedure proves to be a good method to estimate the value of  $D$ , which is important for the quantitative description of autowave processes in excitable media. To achieve high accuracy of the estimate we need to wait about 3 min to be sure that the stationary wave shape is established. In the closed BZ system that we used the propagating velocity decreases during this time by about 0.5%. To improve the accuracy of the estimate it will be helpful to use an open gel reactor for the BZ system [16] in future studies.

The kinematical consideration is also applicable to the case of smooth inhomogeneities as considered, for in-

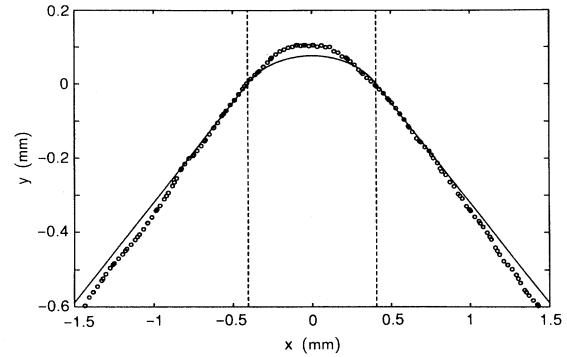


FIG. 6. Shape of the central part of an autowave around the dark stripe observed in experiment (open circles) and calculated by Eq. (4) (solid line). Stripe half width  $W=0.41$  mm. Boundaries of the stripe are indicated by vertical dashed lines.

stance, in Ref. [17]. But in such a case it is necessary to obtain detailed information about the value of the planar wave velocity in all parts of the excitable medium. In our case of the velocity jump, the main reason for existence of the observed stationary patterns is the dependence of normal propagation velocity on local curvature of the wave front. Since curvature-related effects are a common feature of autowaves, similar phenomena should be also a specific property of other types of excitable systems in physics and biology.

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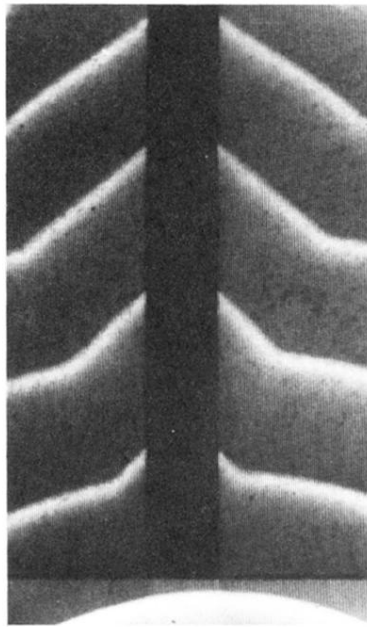


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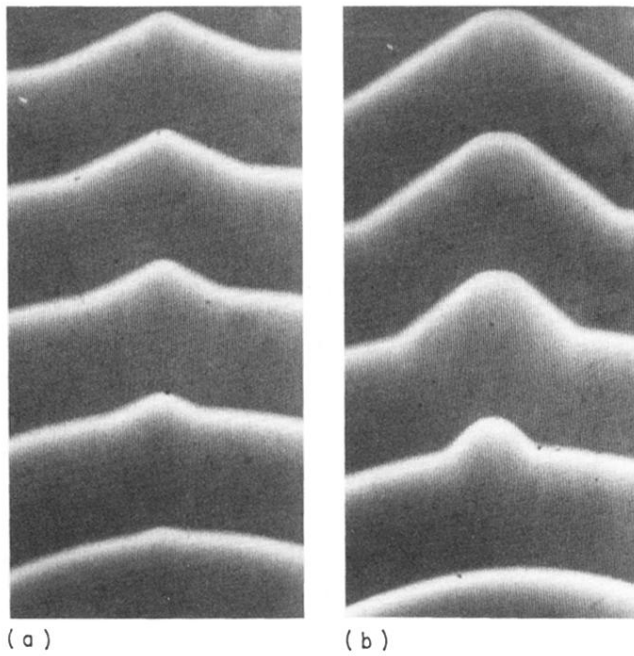


FIG. 2. Shapes of autowaves observed immediately after removing the dark stripes of different half width. Half width  $W$  and observed angle  $\beta_W$ : (a)  $W=0.28$  mm,  $\beta_W=-26.4^\circ$ ; (b)  $W=0.83$  mm,  $\beta_W=-35.7^\circ$ .