Frequency-modulated autowaves in excitable media

A. P. Muñuzuri,[1,*] V. A. Davydov,[2] M. Gómez-Gesteira,[1] V. Pérez-Muñuzuri,[1] and V. Pérez-Villar[1]
[1]*Nonlinear Physics Group, Facultad de Físicas, Universitá de Santiago de Compostela, 15706 Santiago de Compostela, Spain*
[2]*Department of Physics, MIREA, 117454 Vernadskogo 78, Moscow, Russia*
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Refraction of a train of autowaves on a moving boundary, separating two active media of different excitabilities, is studied using the light-sensitive Belousov-Zhabotinsky reaction. It was found that the frequency of the outgoing waves can be smoothly modulated by changing the velocity of the moving boundary. Results are compared with theoretical predictions showing a perfect agreement. They are valid for both autowaves and classical conservative waves. [S1063-651X(96)50912-8]

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Nonlinear active chemical systems, like the Belousov-Zhabotinsky (BZ) reaction [1], are often studied because of the properties of the propagating waves. These waves (autowaves) are also responsible for many processes in nature from living systems to many physical, ecological, and biochemical systems [2]. This multiplicity of applications of the results obtained in chemical systems motivated their exhaustive study in the last years. One of the most interesting applications, perhaps because of the connotations for human life, is the understanding of morphogenesis and functional aspects in some biological systems [2] (e.g., the existence of spiral waves in the cardiac tissue, responsible for some pathologies; propagation failure of signals in nerves, etc. [3,4]) and how these processes can be influenced and/or controlled (mechanisms for controlling and annihilating spiral waves have been developed in the framework of chemical excitable media [5–11]). More recently, autowaves in excitable media were used for image processing [12,13] or, in general, information processing.

In this paper, we present a mechanism of the controlled and smooth variation of the frequency of autowaves. The refraction of autowaves on a static boundary has already been studied both experimentally [14] and theoretically [15,16], finding the Snell's law for autowaves. These properties have been used in the context of catalytic surface reactions [17] and in the BZ reaction where some *V*-shaped structures were created and applied to estimate the diffusion coefficient of the propagator species [18].

The experimental study of this phenomenon was done by using the BZ reaction, catalyzed by the ruthenium bipyridil complex $Ru(bpy)_3^{2+}$ [19,20]. Light induces the release of Br^-, which decreases the velocity of propagation of autowaves. To avoid convection, the catalyst was immobilized in a silica-gel matrix in a Petri dish (gel thickness: 1 mm, preparation as in Ref. [21]; diameter of the Petri dish: 7.1 cm). The solution (0.18*M* NaBr, 0.33*M* malonic acid, 0.39*M* $NaBrO_3$, and 0.69*M* H_2SO_4) was pored into the gel. (Temperature of $25\pm1\,°C$.) White light, leaving horizontally from a 250 W halogen lamp was reflected by a mirror and then reached the reagent. Recording was done with a vertically placed video camera via an interference filter (450.6 nm).

A black spot was placed in one side of the Petri dish, while the rest of the medium was homogeneously illuminated. With the concentrations of the reactants used, the dark region started emitting periodical waves (because the dynamics of the medium in this region is oscillatory). The period of the wave train (67 s) was longer than the spiral one (about 40 s) so that consecutive waves did not interact [22,23]. In the rest of the medium no wave was excited spontaneously (its dynamics is excitable) and just waves coming from the black spot are allowed to propagate through.

Between the Petri dish and the light source, a continuously moving film was placed whose velocity could be controlled. This film consists of a dark part (but not completely opaque, so that the amount of light reaching the dish is diminished but visual measurements are still possible) followed, via a sharp change, by a transparent region. In this way, it is possible to create two different media, with different autowave propagation velocities (the propagation velocity depends on the light intensity), separated by a boundary whose position is changed in time with constant velocity.

The experiments carried out with this setup are exemplified by the sequence of pictures shown in Fig. 1. Circular waves were created on the left-hand side of the Petri dish. The moving boundary separating a dark region on the left-hand side of the Petri dish (high excitability and high propagation velocity, v_1 in the figure) from an illuminated region on the right-hand side (low excitability and low propagation velocity, v_2 in the figure) was coming from the right side of the Petri dish (with velocity u). Figure 1(a) is a picture of the active medium taken when the moving boundary between the enlighten region and the dark part is coming into the observation zone; the most of the medium is still in darkness. During the next 350 s the boundary moved with constant velocity until reaching the position shown in Fig. 1(b) (now the most of the medium is illuminated). Note that the wavelength on the illuminated part is shorter (about 25%) than that in the dark zone. The whole experiment is summarized in Fig. 1(c). This is a vertical array of picture slices, taken every 5 s [slice area: 16.8 mm long, 0.2 mm wide, corresponding with the white line in pictures 1(a) and 1(b)]. Each slice is perpendicular to the moving boundary that, in the

*Electronic address: uscfmapm@cesga.es

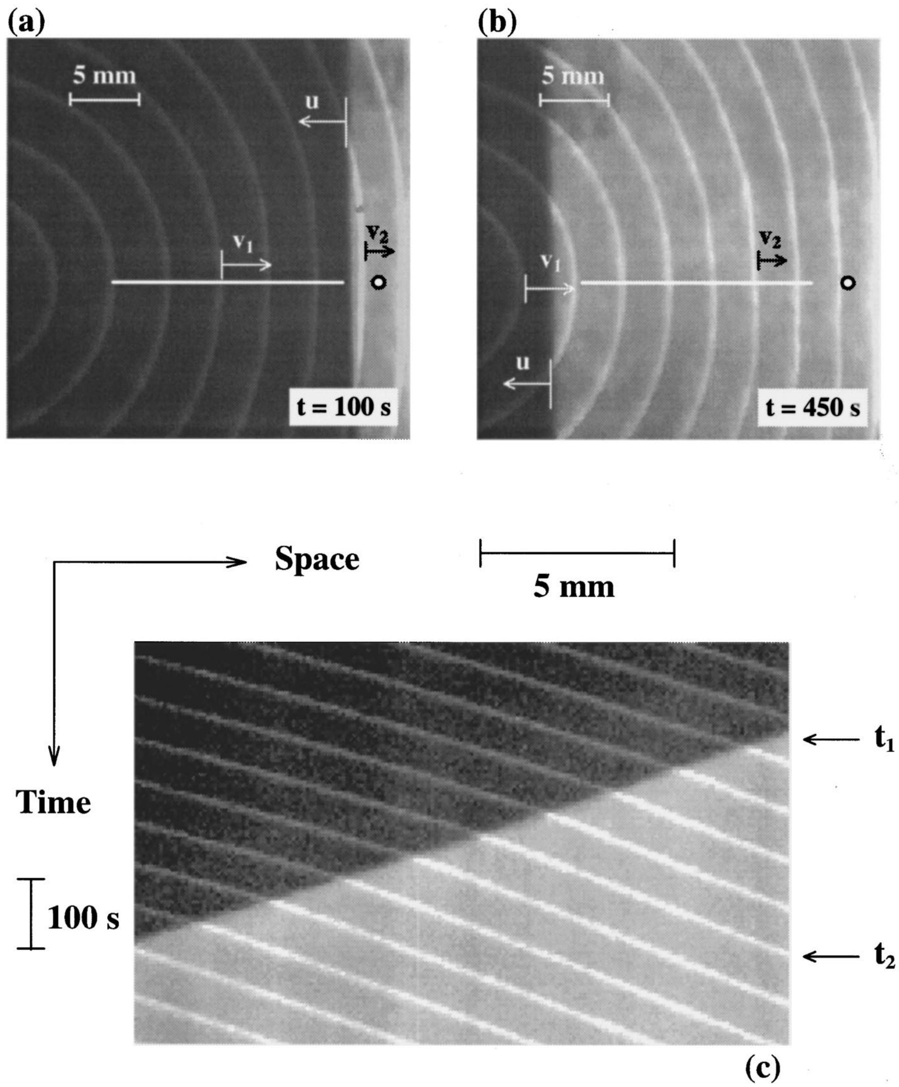

FIG. 1. (a) Picture of the medium 100 s after the beginning of the experiment. The waves are produced by the nonilluminated part of the medium. The moving boundary, separating two parts of the medium with different wave velocities v_1 and v_2, is reaching the observation zone with a constant velocity u. After 350 s, the boundary has moved as shown in (b). (c) Space-time plot of a train of autowaves moving from a dark region to an illuminated one, the boundary between both media also moving (from right to left in the figure with a velocity $u=3.3$ mm/min) in the opposite direction of the autowaves (light stripes in the figure, moving from left to right). $t_1=125$ s, $t_2=427.5$ s.

figure, comes from the right-hand side while the waves, corresponding with the illuminated stripes, are coming from the left-hand side. First, the medium is completely in darkness (just the minimum light intensity was allowed in order to make visual observations) (from $t=0$ to $t=t_1$); then, the boundary reaches the Petri dish [Fig. 1(a)] and, slowly, moves toward the left until the whole Petri dish is illuminated [$t=t_2$, Fig. 1(b)]. Note that when the waves reach the boundary, their velocity is dramatically diminished (just notice the different slope of the lighter stripes).

The temporal evolution of the frequency of autowaves, measured at the right part of the Petri dish [the farthest point to the pulsing spot, marked with a white spot in Figs. 1(a) and 1(b) so that waves are almost planar], is presented in Fig. 2(a). Three different regimes are observed in this figure. First, from $t=0$ to $t=t_1$, no effects were observed; the medium is completely in darkness and the frequency remains constant. Second, the moving boundary is between the measuring point and the pulsing spot ($t_1<t<t_2$); the frequency is dramatically diminished and it remains there until the whole medium is illuminated. After that, a transient regime appears where the frequency slowly recovers its resting value at time t_3. Numerical simulations with a two variable reaction-diffusion (Oregonator) model [24] were performed [see Fig. 2(b)] and the results fit well with the experiments.

It is possible to derive a simple theory that recovers the main observed features. Let us consider a periodic train of autowave pulses propagating in a two-dimensional excitable medium. Let us suppose that the pulses cross a flat penetrable boundary (perpendicular to the OX in the Cartesian frame of reference) between two excitable media with differences in diffusivities and in local kinetics. Neglecting the possible interaction between propagating waves (refractoriness), we can consider the velocities of the plane fronts in both media to be constant and equal to v_1 (on the left-hand side of the medium) and v_2 (on the right-hand side of the boundary), as shown in the experimental Fig. 1.

If the boundary is fixed, only the wave vector of the crossing train changes but the frequency remains invariable. However, if the boundary moves with some nonzero velocity u, the frequency will be also changed. Let us consider this variation. The dependence of the U variable on the (x,y) coordinates and time, t, can be described in each side of the medium by

$$\begin{aligned} U_1&=F_1(K_{1x}x+K_{1y}y-\omega_1 t),\\ U_2&=F_2(K_{2x}x+K_{2y}y-\omega_2 t), \end{aligned} \tag{1}$$

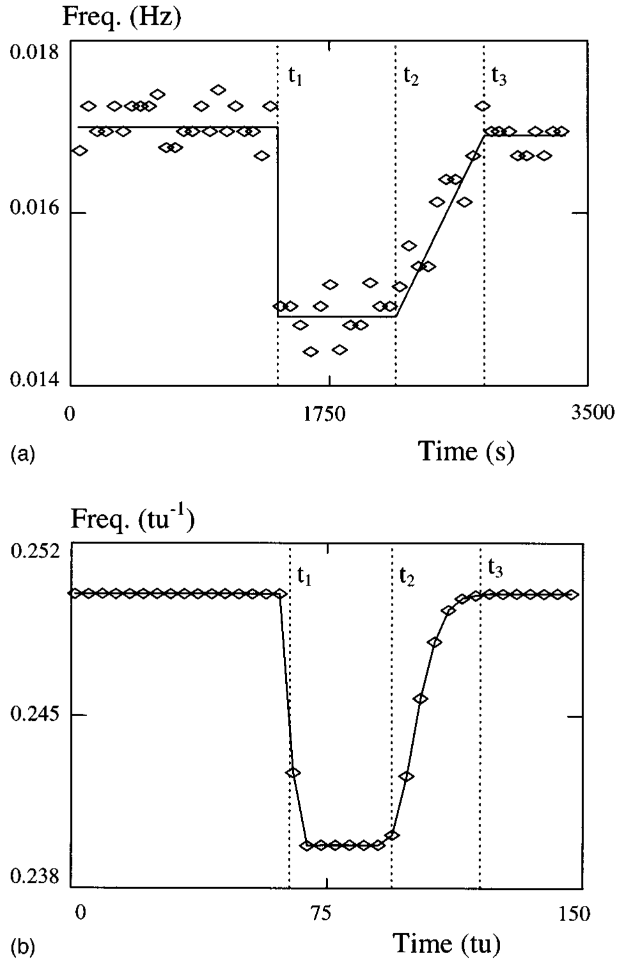

FIG. 2. Temporal evolution of the frequency of the autowaves, measured at the right of the Petri dish (opposite side of the pulsing spot). (a) Results corresponding to the experiment shown in Fig. 1. Solid line is just a guide for eyes; $t_1=1400$ s, $t_2=2200$ s, $t_3=2800$ s. (b) Numerical results with the two-variable Oregonator model modified to account for the light effect. $t_1=64$ tu, $t_2=94$ tu, $t_3=120$ tu. Model parameters: $f=3$, $q=0.002$, $\varepsilon=0.01$, light intensity at the beginning, dark region, $\varphi=0$, and at the end, illuminated region, $\phi=0.002$ and boundary velocity $u=9.6$ su/tu (su and tu denote space and time units, respectively).

where F_1 and F_2 are 2π-periodic functions. K_{1x}, K_{1y}, K_{2x}, K_{2y} are projections of the wave vector and ω_1, ω_2 are the frequencies of the train of pulses in both media. U_1 and U_2 are the activator variables in both sides of the moving boundary.

Let us suppose that the boundary moves toward the negative part of the OX axis. On the boundary $(x=-ut)$, the concentrations U_1 and U_2 must be equal in any point y and at any instant t. These conditions are satisfied only if

$$K_{1x}u+\omega_1=K_{2x}u+\omega_2,\qquad K_{1y}=K_{2y}, \tag{2}$$

or

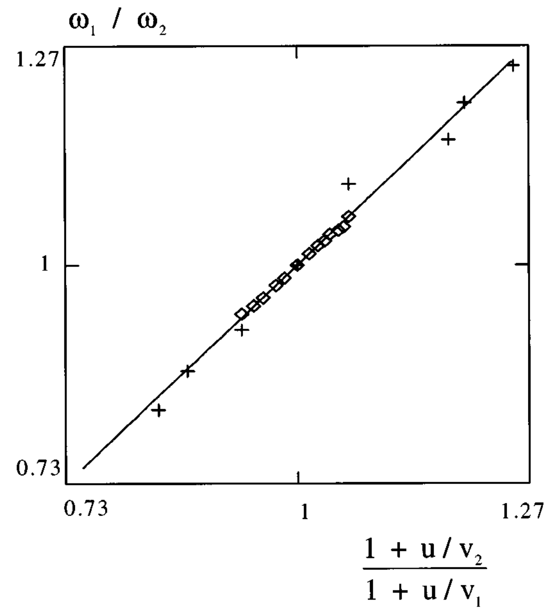

FIG. 3. Dependence of ω_1/ω_2 on $(1+u/v_2)/(1+u/v_1)$. The continuous line is the theoretical prediction, crosses are the experimental points, and the rhombi the numerical results. Different points correspond to experiments with different boundary velocities and light intensities.

$$\omega_1+\omega_1\frac{u}{v_1}\cos\alpha_1=\omega_2+\omega_2\frac{u}{v_2}\cos\alpha_2,\qquad \frac{\omega_1}{v_1}\sin\alpha_1=\frac{\omega_2}{v_2}\sin\alpha_2, \tag{3}$$

where α_1 is the incident angle and α_2 is the angle of refraction.

For the trivial case, $u=0$, we have $\omega_1=\omega_2$ and $\sin\alpha_2=(v_2/v_1)\sin\alpha_1$ (Snell's law for autowaves [15]). For a nonzero value of the boundary velocity, u, Eqs. (3) describe the refraction of autowaves on a moving boundary (α_2 as a function of α_1) as well as a change in the frequency. Thus, the refraction law is

$$\cos\alpha_2=\frac{\sqrt{v_2^2u^2+(f^2+u^2)(f^2-v_2^2)}-v_2u}{f^2+u^2}, \tag{4}$$

where

$$f=\frac{v_1+u\cos\alpha_1}{\sin\alpha_1}. \tag{5}$$

Substituting Eqs. (4) and (5) into Eq. (3), it is possible to obtain the expression for ω_2 as a function of ω_1 and α_1. This expression essentially gets simplified in the case of normal incidence $\alpha_1=0$,

$$\omega_2=\omega_1\frac{1+u/v_1}{1+u/v_2}. \tag{6}$$

Note that $\omega_2>\omega_1$ if $v_2>v_1$ and vice versa. Equation (6) was obtained for the case when the boundary moves to meet the autowave pulses. A change in the sign of the velocity u can also be considered and the results become slightly more complicated [25].

Equation (6) is compared with experimental and numerical measurements in Fig. 3. Different experiments were done for different values of u and light intensities and the results are shown in Fig. 3 where the value of ω_1/ω_2 is plotted as a function of $(1+u/v_2)/(1+u/v_1)$. Note that both experimental (crosses in the figure) and numerical (rhombi in the figure) results fit well to the straight line predicted theoretically.

Equation (6) is valid for classical waves so that the results here presented are generic for wave propagation. This turns to be untrue for the autowaves case when the distance between consecutive waves becomes shorter. Now, they start interacting because of the temporal inhibition of the medium after the passage of an autowave which is not the case of classical waves. The result is that the frequency shift introduced by the moving boundary on the refracted waves is diminished. This mechanism for smooth control of the frequency of autowaves, by variation of the boundary velocity, also points out a way of encrypting or encoding information in a train of autowaves, just by changing the direction of motion and velocity of the boundary.

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