

Tracking Waves and Vortex Nucleation in Excitable Systems with Anomalous Dispersion

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We report experimental results obtained from a chemical reaction-diffusion system in which wave propagation is limited to a finite band of wavelengths and in which no solitary pulses exist. Wave patterns increase their size through repeated annihilation events of the frontier pulse that allow the succeeding pulses to advance farther. A related type of wave dynamics involves a stable but slow frontier pulse that annihilates subsequent waves in front-to-back collisions. These so-called merging dynamics give rise to an unexpected form of spiral wave nucleation. All of these phenomena are reproduced by a simple, three-species reaction-diffusion model that reveals the importance of the underlying anomalous dispersion relation.

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Propagating waves of excitation are known to exist in numerous biophysical and physicochemical systems [1,2]. Important examples include neuronal and cardiac tissue as well as gas discharge systems and chemical reaction-diffusion (RD) media. The dynamics of wave trains are dominated by the dispersion relation which relates the pulse speed c to the period T . Most experimental systems show a monotonically increasing dispersion relation $c(T)$ that converges to the velocity of a solitary pulse for very large periods [3]. Moreover, one finds a low-period limit below which no stable wave trains exist. Exceptions to this normal scenario are found in the catalytic reduction of NO with CO on Pt(100) surfaces [4], the 1,4-cyclohexanedione Belousov-Zhabotinsky (CHD-BZ) reaction [5,6], in neuronal tissue [7], and perhaps in other biological systems [8]. In these cases, the dispersion relation shows nonmonotonic behavior, but nonetheless extends to infinitely large periods.

Nonmonotonic dispersion relations give rise to several remarkable phenomena that are not found in systems with normal dispersion, such as nonunique rotation periods of spiral waves [9], stable pulse multiplets [10], unusual steady-state solutions of front lines [11], and different velocities for the same wavelength [12]. In this context, the CHD-BZ reaction has become a valuable model. It involves the oxidation of 1,4-cyclohexanedione by bromate in an acidic medium. Similarly to the classic malonic acid-BZ reaction [1], the main nonlinearity in the CHD-BZ system stems from the autocatalytic production of bromous acid, but it does not induce the formation of undesired gas bubbles. In this Letter we describe an unexpected low-frequency limit for pulse trains in the CHD-BZ systems that gives rise to a tracking-like evolution of wave patterns. Moreover, we identify a novel nucleation mechanism of spiral waves. These results are reproduced by a simple, three-species RD model.

Our simulations are based on Barkley's model [13] which involves a fast activator u and a slow inhibitor variable v . We extend this model by introducing a second

inhibitory control species w according to

$$\frac{\partial u}{\partial t} = \nabla^2 u + \frac{1}{\epsilon} \left\{ u(1-u) \left(u - \frac{v+w}{a} \right) \right\}, \quad (1)$$

$$\frac{\partial v}{\partial t} = u - v, \quad (2)$$

$$\frac{\partial w}{\partial t} = \beta(\delta - w) - \gamma uw, \quad (3)$$

where ϵ , a , β , γ , and δ are dimensionless constants. Figure 1(a) shows a typical example of a solitary excitation wave propagating in a one-dimensional system towards the right. In the back of the leading u pulse, the system is refractory due to high values of v . Moreover, w decreases during the phase of high u values and slowly relaxes back to its steady-state value δ . Since v and w represent the concentrations of inhibitory species, this scenario creates a nonmonotonic inhibition profile in the wake of the pulse.

To investigate the dispersion relation of our model, we study the rotation period of single pulses propagating in circular, one-dimensional media. This approach mimics an infinite wave train while avoiding wave train instabilities that could give rise to nonuniform interpulse distances. Figure 1(b) shows the resulting pulse speed as a function of the rotation period T for different values of δ . The four dispersion relations are clearly anomalous and show a single maximum as well as a high-frequency limit. As the value of δ is increased, the low-frequency tail of the dispersion relations is lowered in accordance with the expected δ -controlled inhibition far from the pulse. Moreover, the upper two curves show a unique period T_0 for which $c(T_0) = c_0$ and $dc/dT > 0$. This period corresponds to the interpulse period in stable stacked pulse clusters. For $\delta = 0.28$, no such stable period exists and trailing pulses annihilate in the back of the slow leading front. For even larger values of δ (here 0.30), the tail of the dispersion relation becomes unstable giving

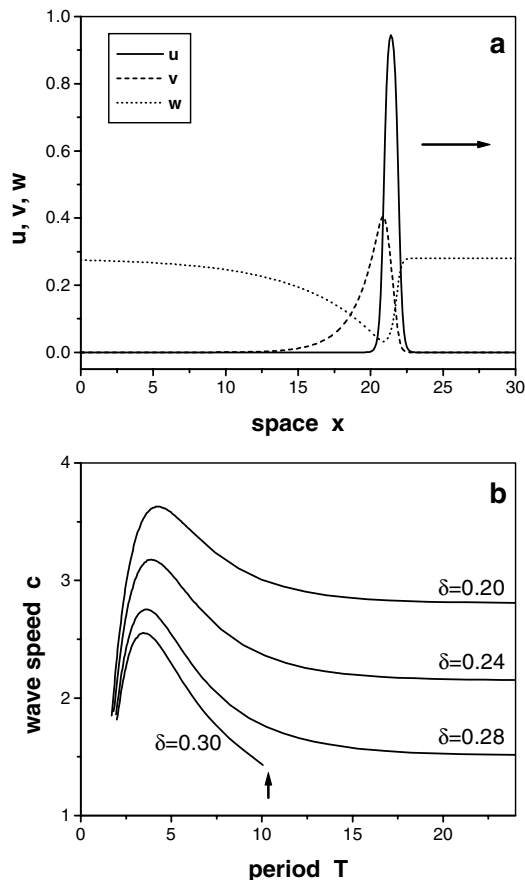


FIG. 1. (a) Solitary wave pulse in a three-species model of an excitable RD systems for $\delta = 0.28$. (b) Four representative dispersion relations that correspond to stacking ($\delta = 0.20$, 0.24), merging ($\delta = 0.28$), and tracking ($\delta = 0.30$) waves. The lowest curve is confined to a finite band of periods. The other parameters are as follows: $\epsilon = 10^{-2}$, $a = 0.7$, $\beta = 0.3$, and $\gamma = 5.0$.

rise to a finite band of allowed interpulse periods and wavelengths. Notice that this phenomenon shares certain similarities with "packet" waves in BZ micro-emulsions [14].

The wave evolution in one-dimensional systems without periodic boundary conditions is illustrated in Fig. 2. Each picture represents a space-time plot in which time evolves in upward direction. Frames (a)–(c) are obtained through numerical integration of Eqs. (1)–(3), whereas frames (d)–(f) are experimental data. In our simulations, excitation pulses are initiated at the left boundary by periodic perturbations of the system. The experiments are carried out by filling thin capillary tubes (inner diameter 0.8 mm, length 90 mm) with the CHD-BZ solution.

Stock solutions of CHD (Aldrich), NaBrO_3 (Fluka), and H_2SO_4 (Riedel–de Haën) are prepared in nanopure water. As the redox catalyst we use $\text{Fe}[\text{batho}(\text{SO}_3)_2]_3^{4-}$ (25 mM) because of its high contrast between the reduced and oxidized state [15]. Very similar results are also obtained with $\text{Fe}(\text{phen})_3^{2+/3+}$ (ferroin/ferriin) at smaller

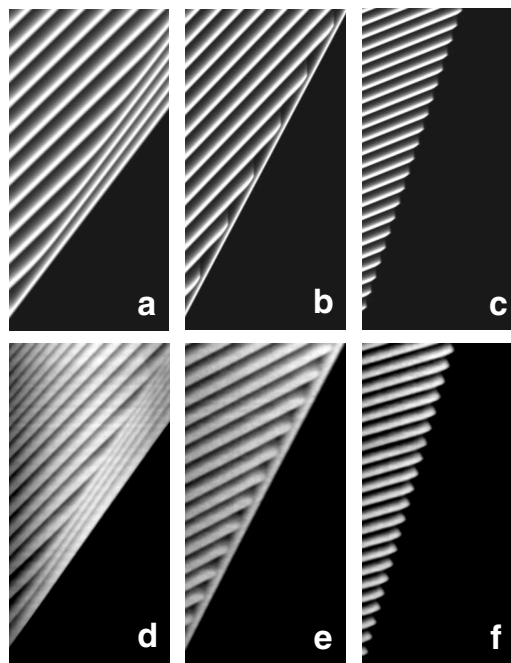


FIG. 2. Space-time plots of stacking, merging, and tracking waves. (a)–(c) Numerical simulations with $\epsilon = 10^{-2}$, $a = 0.7$, $\beta = 0.3$, $\gamma = 5.0$, and $\delta = 0.24$ (a), 0.28 (b), 0.30 (c). (d)–(f) Experimental data for the initial concentrations in mol/L: $[\text{H}_2\text{SO}_4] = 0.6$, $[\text{CHD}] = 0.2$, $[\text{Fe}[\text{batho}(\text{SO}_3)_2]_3^{4-}] = 0.0005$, and $[\text{NaBrO}_3] = 0.30$ (d), 0.25 (e), 0.20 (f). The horizontal and vertical axes span 26.8 mm (d), 7.1 mm (e), 5.7 mm (f), and 375 s (d), 300 s (e), 600 s (f), respectively.

$[\text{NaBrO}_3]$. The reaction system is monitored with a charge-coupled-device camera that resolves concentration differences between the reduced and oxidized forms of the involved catalyst. Accordingly, oxidation waves appear as bright bands in Figs. 2(d)–2(f). In the experiments, waves nucleate spontaneously at the open ends of the capillary tube of which only the left side is shown. At these ends bromine gas escapes from the solution into the atmosphere, thus, shifting the medium locally into an oscillatory state. The resulting initiation period remains essentially constant in time, but varies between different experimental runs.

Figures 2(a) and 2(d) show typical examples of stacking waves. Here, fast excitation fronts approach the back of a leading, slow pulse and form closely spaced wave packets. The dispersion relation has the main features of the curves shown in Fig. 1(b) for $\delta = 0.20$ and 0.24 . Accordingly, the speed and the period within the wave packets corresponds to c_0 and T_0 . The dynamics in Figs. 2(b) and 2(e) are caused by merging waves that obey dispersion relations similar to the one shown in Fig. 1(b) for $\delta = 0.28$. Finally, Figs. 2(c) and 2(f) present examples for tracking waves. In this dynamic regime, the first wave pulse travels only a short distance before vanishing in the highly inhibited medium. Subsequent pulses, however, travel an increasingly longer distance since their predecessors have partially "cleared the way"

by decreasing the local concentrations of inhibitory species. This behavior is characteristic for excitable systems with finite-bandwidth dispersion relations (cf., Fig. 1(b), $\delta = 0.30$). To the best of our knowledge, this is the first example for this interesting and unusual form of wave propagation. Moreover, we find in all three cases an excellent qualitative agreement between experiments and simulations.

In the tracking examples shown in Figs. 2(c) and 2(f) the period of initiation was chosen to be on the stable branch (i.e., $dc/dT > 0$) of the finite bandwidth dispersion relation. This assures that the overall wave pattern grows at a constant speed C_t which depends on the wavelength of the wave train λ_i , the decay length Λ_i of pulses entering the steady-state medium at the leading edge of the pattern, as well as on their average speed C_i during the decay. Assuming that C_i is approximately the wave velocity c_i within the pulse train, we obtain a growth speed of

$$C_t = c_i \frac{\Lambda_i}{\Lambda_i + \lambda_i}. \quad (4)$$

Analysis of the experimental data in Fig. 2(f) yields $\lambda_i = 1.7$ mm, $\Lambda_i = 0.13$ mm, $c_i = 80$ $\mu\text{m/s}$, and $C_t = 5.4$ $\mu\text{m/s}$, which is in good agreement with the value of $C_t = 5.7$ $\mu\text{m/s}$ predicted by Eq. (2).

However, tracking behavior can be significantly more complex than the dynamics shown in Figs. 2(c) and 2(f). If the initiation period is on the unstable branch ($dc/dT < 0$) of the dispersion relation, we observe non-uniform growth dynamics that involve phases of expansion as well as phases of sudden retraction. Consequently, the boundary of the wave pattern has a very rugged and complicated shape in the space-time plots suggesting that Λ_i and C_i depend on λ_i for wave trains with long wavelengths. This behavior is related to the transient bunching dynamics of stacking wave trains, in which an unstable wave packet decays into nearly stable clusters with closely stacked pulses that are spaced at long, and hence least unstable, intercluster distances [6]. Whereas this cascade of bunching events will eventually lead to one large cluster with uniform pulse spacing, it is currently unclear how, if at all, tracking waves with $dc_i/dT_i < 0$ establish a stationary state.

Stacking, merging, and tracking waves also exist in two-dimensional systems [16]. To create pseudo-two-dimensional conditions in our experiments, we typically confine thin layers of the reaction solution between two glass plates spaced at 0.5 mm. In the following, we discuss a novel mechanism for spiral wave nucleation as a highly unexpected example of the wealth of phenomena in excitable systems with anomalous dispersion.

Figure 3 shows four consecutive snapshots of wave propagation in the CHD-BZ reaction. In this particular experiment, a target pattern of concentric wave rings forms around a small inhomogeneity. To its upper right,

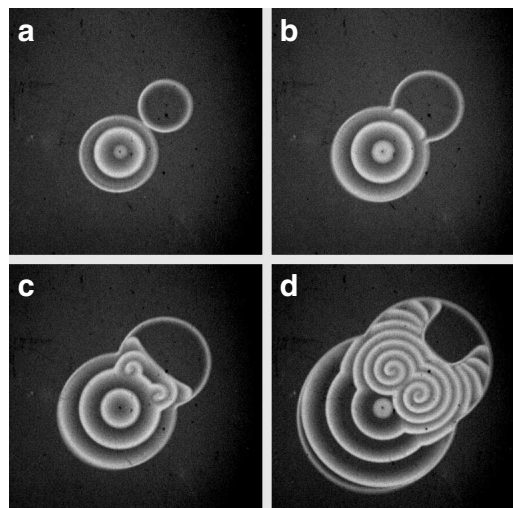


FIG. 3. Consecutive snapshots of merging waves in a thin layer of the CHD-BZ reaction. A pair of spiral waves nucleates from the second wave front emitted by the target pattern. Initial concentrations are the same as in Fig. 2(e). Image area: 6.9 cm². Time between frames: 27, 33, and 70 s.

a second wave is initiated. This process can occur spontaneously or it can be induced in a controlled fashion via electrochemical initiation. The reaction medium in Fig. 3 creates waves that show merging behavior similar to the one-dimensional examples in Figs. 2(b) and 2(e). The occurrence of merging events can be seen most clearly along the lower, left edge of the wave structure in Fig. 3(d). As the outermost circular fronts collide [Fig. 3(a)], they undergo mutual annihilation as expected for typical excitation waves. However, the second wave front emitted by the pacemaker of the target pattern has no counterpart to collide with. Moreover, this second pulse travels faster than the, now fused, 8-shaped leading front. This velocity difference culminates in a front-to-back collision in which the trailing front vanishes. This merging process is interrupted along a short, central segment where the trailing pulse survives, thus entering a wave-free circular domain left behind by the upper, solitary front. The resulting banana-shaped front segment has a nearly perpendicular orientation with respect to the leading front [Fig. 3(b)]. This scenario induces the formation of a pair of counter-rotating spiral waves [Fig. 3(c)] that eventually becomes the primary pacemaker of the overall wave pattern since their rotation frequency is greater than the frequency of the target pattern.

To evaluate whether the vortex nucleation mechanism in Fig. 3 can be reproduced by our model, we carried out numerical simulations that employ initial conditions very similar to those of the experiment. Figure 4 shows a sequence of six snapshots of the variable $v(x, y, t)$. The simulations involve two initiation sites along the $y = x$ diagonal of the two-dimensional system. One site acts as a periodic pacemaker, whereas the other one triggers only one wave pulse [Fig. 3(a)]. The evolution of this system

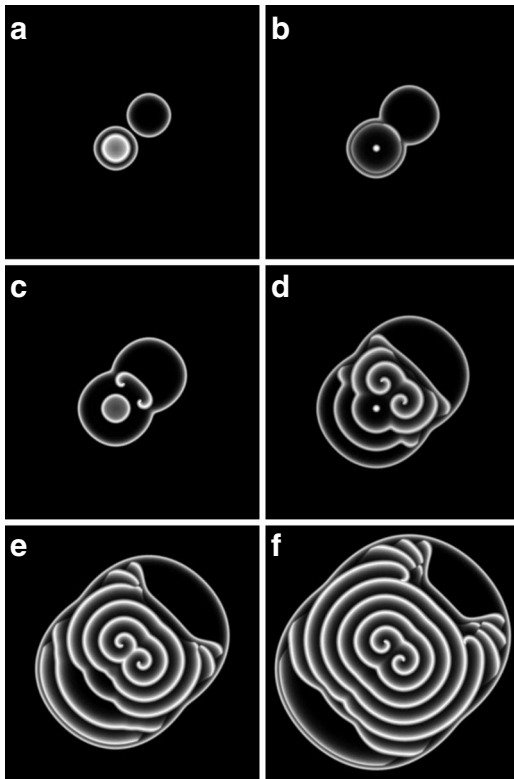


FIG. 4. Consecutive snapshots of merging waves and spiral nucleation obtained by numerical integration of Eqs. (1)–(3). Model parameters: $\epsilon = 10^{-2}$, $a = 0.8$, $\beta = 0.3$, $\gamma = 5.0$, and $\delta = 0.28$.

shows excellent agreement with the experimental data as it induces the formation of a single pair of spiral waves. Moreover, our simulations successfully reproduce the u-like shape of the second front in the upper left quadrant. This peculiar shape stems from the interplay of curvature effects and the nonmonotonic inhibition profile left behind by the leading front.

Last, we suggest that the spiral nucleation mechanism described in this Letter is the primary source of spirals that form in large numbers in CHD-BZ systems with merging waves. While the specifics depend strongly on the density, frequency, and relative location of the initial pacemakers, it is likely that nearly all spirals result from processes, the main essentials of which are captured by the simple scenario shown in Figs. 3 and 4. This mechanism is, therefore, important and characteristic for excitable systems with anomalous dispersion that lack a stable fixed point $c = c_0$. Moreover, it should be distinguished from other instabilities that generate vortices through spiral breakup [17] or after external wave initiation in the refractory zone of an excitation pulse [18].

In conclusion, we have devised and tested a simple, three-species RD model that captures all of the phenomena currently known to exist in the CHD-BZ reaction. These include stacking and merging wave trains that result from nonmonotonic, nonoscillatory dispersion re-

lations. Moreover, for media with a highly inhibited rest state, we discovered finite-bandwidth dispersion relations that induce tracking wave patterns. The latter structures expand despite the lack of stable, solitary fronts. For merging waves, we identified a novel mechanism of spiral wave generation from simple, nonrandom initial conditions. It will be interesting to explore whether these phenomena can be found in biological systems that obey anomalous dispersion relations.

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