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## Bloch-front turbulence: theory and experiments

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### Abstract

We report on experimental and theoretical findings of front destabilization that causes spontaneous spiral–vortex nucleation and produces a state of spatio-temporal disorder. The experiments were carried out on an oscillatory photosensitive Belousov–Zhabotinsky reaction that is periodically forced in time. Numerical studies were carried out on a modified Complex Ginzburg–Landau equation and on the FitzHugh–Nagumo model. Using velocity–curvature relations for fronts we associate the onset of spatio-temporal disorder with the Non-equilibrium Ising Bloch (NIB) bifurcation, and study the generic patterns that form on both sides of the bifurcation as the distance from it is increased.

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## 1. Introduction

The onset of spatio-temporal chaos in extended systems often involves the spontaneous appearance of phase singularities in the form of either spiral vortices or defects [1,2]. Various realizations of this phenomenon have been studied both in models and in experiments [3–5]. In most studies the singularities were preceded by instabilities of periodic patterns. Instabilities of localized structures and their impact on pattern formation have also been studied theoretically and experimentally. Transverse instabilities of fronts, for example, have been related to the formation of stationary dendrites [6] and labyrinths [7,8]. Very few studies, however, addressed the possibility of a front instability leading to phase singularities and spatio-temporal chaos.

In this paper we present theoretical and experimental evidence for a state of spatio-temporal chaos in bistable systems that is maintained by repeated nucleation of spiral–vortex pairs along front lines. The state, which we call “Bloch-front turbulence”, occurs in the vicinity of the Nonequilibrium Ising Bloch (NIB) front bifurcation. The NIB bifurcation is a pitchfork bifurcation from a stationary “Ising” front to a pair of counter-propagating “Bloch” fronts as a system control parameter is varied. It has been analyzed in the FitzHugh–Nagumo (FHN) [9] and Complex Ginzburg–Landau (CGL) equations [10], and has been observed in liquid crystals [11,12], chemical reactions and catalytic surface reactions [13]. In addition, we further study the patterns that form on both sides of the NIB bifurcation as the distance from the bifurcation is increased. The results reported here extend earlier theoretical and experimental results [14].

## 2. Spiral–vortex nucleation

We first present experimental results demonstrating spontaneous vortex nucleation along front lines and the subsequent development of Bloch-front turbulence. The experiments were carried out on an oscillatory photosensitive Belousov–Zhabotinsky (BZ) reaction that is periodically forced in time with spatially uniform light pulses [15]. Details about the experimental setup can be found in Refs. [15,16]. The forcing frequency is chosen to be approximately twice the uniform oscillation frequency of the unforced reaction. Under this condition the system is bistable with two coexisting stable states of uniform oscillations at half the forcing frequency and with oscillation phases differing by  $\pi$ . Fig. 1(left) shows the time evolution of an initial planar front between the two uniform states and the spontaneous nucleation of spiral–vortex pairs along it. As the nucleation process continues, a disordered state occupying the whole physical domain develops. In the asymptotic disordered state the vortices are sufficiently dense for the rate of vortex annihilation to become comparable to that of vortex nucleation. As a result, the number of vortices fluctuates around a mean value.

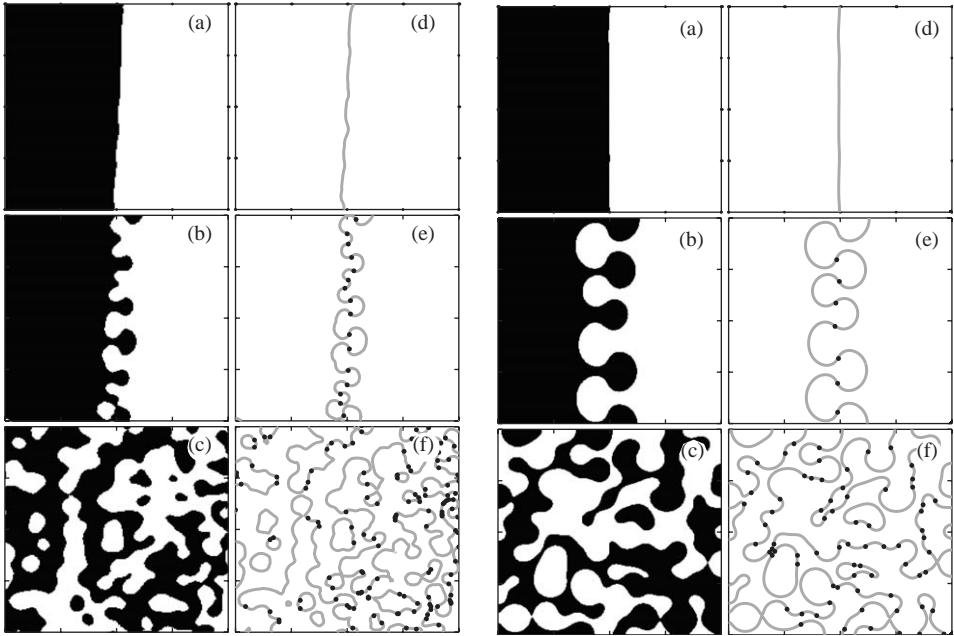


Fig. 1. Spiral vortex nucleation in the BZ system (left) and in the CGL equations (right). Frames (a)–(c) show the phase of the oscillations at near half the driving frequency at three successive times. Frames (d)–(f) show the position of the vortices along the front at the corresponding times. (a,d) The initial nearly planar front is unstable to transverse perturbations. (b,e) Vortices form in pairs along the front. (c,f) Vortices eventually fill up the entire system. More details can be found in Ref. [14].

Similar processes have been found in numerical solutions of CGL equations (with a term accounting for the periodic forcing [14]) and of the FHN model near the NIB bifurcations [17]. Fig. 1(right) shows, in the CGL equations, the destabilization of a planar front followed by the spontaneous nucleation of vortex pairs and the convergence to a disordered state of Bloch-front turbulence.

### 3. Pattern formation and the NIB bifurcation

By changing a control parameter to cross the NIB bifurcation, the following main sequence of patterns is observed. Starting in the Bloch regime (small forcing amplitudes in the experiment and in the CGL equations) stable spiral waves are observed. As the NIB is crossed, the spirals become unstable and we find Bloch-front turbulence. Moving away from the NIB bifurcation into the Ising regime (higher forcing amplitudes) labyrinthine patterns develop. Farther into the Ising regime large domain patterns are found. The complete sequence of patterns observed in the experiment while changing parameters across the NIB bifurcation is shown in Fig. 2. The corresponding sequence as obtained by numerical solutions of the FHN model

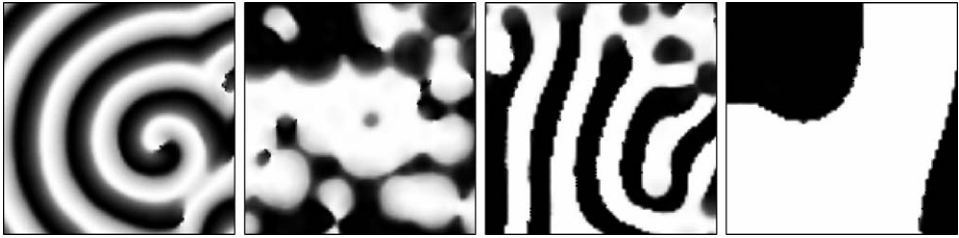


Fig. 2. The transition from rotating spirals to spatio-temporal chaos, labyrinths, and uniform patterns in the BZ system. The forcing frequency is held constant at 0.03 Hz and the forcing intensity is increased slowly from left to right: 32.8, 42.3, 44.8, 53.2 W/m<sup>2</sup>. The frame size is 5.41 mm × 5.41 mm. Chemical conditions: Reservoir A: 0.001 M tris(2,2'-bipyridyl)dichlororuthenium(II) hexahydrate, 0.8 M H<sub>2</sub>SO<sub>4</sub>, 0.184 M KBrO<sub>3</sub>; Reservoir B: 0.32 M malonic acid, 0.3 M NaBr, 0.8 M H<sub>2</sub>SO<sub>4</sub>, 0.184 M KBrO<sub>3</sub>.

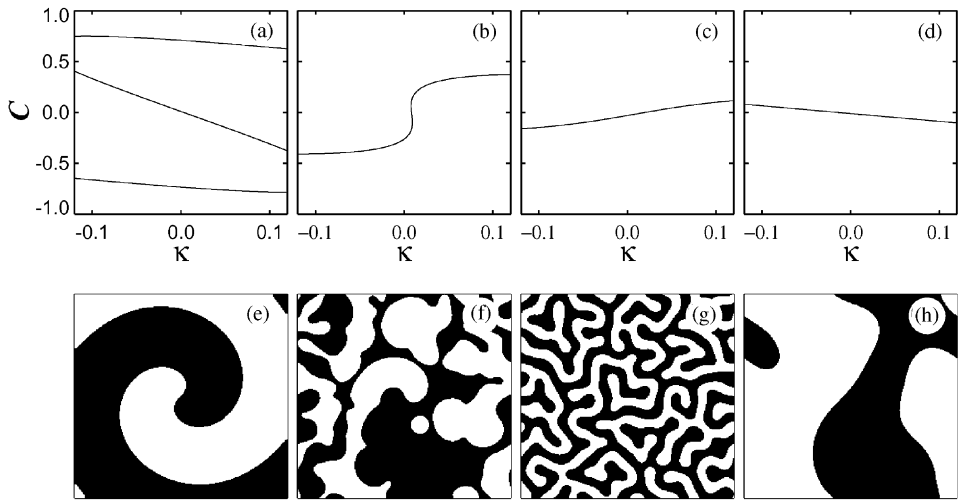


Fig. 3. Top: the relation between the normal front velocity,  $C$ , and front curvature,  $\kappa$ , varying the control parameter  $\varepsilon$  in an FHN reaction–diffusion model (see Footnote 1). (a) Counter-propagating Bloch fronts,  $\varepsilon = 0.01$ , (b) near the NIB bifurcation,  $\varepsilon = 0.035$ , (c) Ising front with transverse front instability,  $\varepsilon = 0.07$ , (d) Ising front with no transverse front instability,  $\varepsilon = 1.0$ . Bottom: two-dimensional pattern types corresponding to the parameters in the top  $C$ – $\kappa$  relations. (e) Spiral wave, (f) Bloch-front turbulence, (g) labyrinthine pattern, (h) large domains of uniform states.

is shown in Fig. 3. The control parameter in the FHN model is the ratio,  $\varepsilon$ , of the activator time scale to the inhibitor time scale [9].<sup>1</sup> The same sequence can also be reproduced by solving the CGL equations.

<sup>1</sup>The FHN model we use is  $u_t = u - u^3 - v + \nabla^2 u$ ,  $v_t = \varepsilon(u - 2v - 0.01) + 2\nabla^2 v$ . The solution domain is a  $256 \times 256$  square.

#### 4. The pattern formation mechanisms

The appearance of spiral waves in the Bloch regime far from the NIB is well understood: the spiral-wave core is a short segment along the front line with a transition from one Bloch front to the other [18,19]. The appearance of labyrinthine patterns in the Ising regime away from the NIB is also fairly well understood. The early evolution of the pattern is driven by a transverse front instability [17]. At later stages, repulsive front interactions become important and are responsible for the final length scale that characterizes the pattern [8,20]. Additionally, in the Ising regime the stabilization of fronts to transverse perturbations causes the existence of large domain patterns. Domains of either uniform state typically either grow or shrink to fill the entire system [21].

To understand the Bloch-front turbulence mechanism and why it is found at an intermediate range in parameter space between spiral waves and labyrinths, we use the relations between the normal velocities of fronts,  $C$ , and their curvatures,  $\kappa$ . Fig. 3 shows four  $C$ - $\kappa$  relations corresponding to the four pattern types shown in Fig. 3: spiral waves, Bloch-front turbulence, labyrinths and large domain patterns. The  $C$ - $\kappa$  relations have been calculated using a singular perturbation analysis of the FHN model for weakly curved fronts [17].

Far from the NIB bifurcation in the Bloch regime (Fig. 3(a)) there are three  $C$ - $\kappa$  branches showing near linearly decreasing dependences of normal velocity on curvature. The middle branch corresponds to the unstable Ising front and the other two branches to the pair of Bloch fronts. The negative slopes imply stability of the Bloch front to transverse perturbations. These conditions give rise to stable spiral waves.

Far from the NIB bifurcation in the Ising regime (Fig. 3(c)) there is a single branch, corresponding to the single Ising front. The positive slope over a wide curvature range implies instability to transverse perturbations of both planar and curved Ising fronts. This instability, together with repulsive front interactions (due to inhibitor diffusion in the FHN model), leads to labyrinthine patterns.

In the vicinity of the NIB bifurcation, the  $C$ - $\kappa$  relation takes the form of the universal unfolding of the pitchfork bifurcation as shown in Fig. 3(b). Two features of this form are significant. The first is that the Bloch front branches terminate at a small absolute curvature values, implying that small realizable curvature values that develop in the course of the pattern evolution may induce a transition to the other Bloch front branch. A transition of that kind, occurring along a finite front segment, involves a reversal in the propagation direction of this segment which is accompanied by the nucleation of a vortex pair. The second significant feature is that the slope of the  $C$ - $\kappa$  relation at small curvature values is positive implying a transverse front instability. This instability generates front segments with negative curvatures that increase in time and provides the driving force for vortex pair nucleation. This is exactly the process shown in Fig. 1.

Finally, much farther into the Ising regime (high  $\varepsilon$  values in the FHN model) the  $C$ - $\kappa$  relation can have a negative slope, as Fig. 3(d) shows, the Ising front becomes stable to transverse perturbations and large domain patterns prevail.

## 5. Conclusion

The results reported here suggest the existence of a generic sequence of states across the NIB bifurcation in bistable systems: Bloch spiral waves, Bloch front turbulence, Ising labyrinths and large domain Ising patterns (see Figs. 2 and 3). The results are based on numerical studies of two different models and on experimental studies of the forced BZ reaction.

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