Experimental Evidence for Zigzag Instability of Solitary Stripes in a Gas Discharge System

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The experimental observation of a zigzag instability of the stripes in a planar dc-driven gas discharge system is reported. The stripes in the spatial distribution of current develop from a homogeneous state as the global current is increased. It is shown that the phenomenon is specific for solitary stripes. This finding confirms recent theoretical predictions related to the instability of these localized states in reaction-diffusion media, which is followed by generation of the traveling zigzag wave propagating along a stripe. [S0031-9007(97)02919-0]

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Stripe (or roll) patterns are know to be among the generic structures which are spontaneously generated in two-dimensional (2D) pattern forming systems [1,2]. Systems with the reaction-diffusion (R-D) mechanism of instability can create both small-amplitude periodic stripe patterns [3] and high-amplitude solitary stripes [4,5], the latter being an example of localized states (LS) of active media. The formation of small-amplitude periodic patterns can be properly discussed within the scope of the weakly nonlinear analysis, in terms of interacting harmonic modes. This interaction also originates secondary instabilities of periodic stripe patterns which bifurcate to zigzag or varicose patterns (see, e.g., Ref. [2]). A localized stripe of a R-D medium represents a band domain with a high concentration of activator embedded on the background with the low activator density. It has been shown theoretically that these LS's can exist in a 2D system at the proper set of parameters [4,5]. Recent analysis of the problem of stability of localized stripe solutions of R-D models shows that they can undergo zigzag or varicose instabilities [4–8]. The theory predicts possibilities of destabilization both to stationary and unstationary zigzag modes. Despite the interest in such further development of the LS's in pattern forming systems, up to now we are not aware of an experimental proof of the existence of this phenomenon.

In the present work we give experimental evidence of zigzag destabilization of localized stripes in a pattern forming system. These stripes are formed in a cryogenic gas discharge device as a result of a sequence of bifurcations from the initially homogeneous state. Essentially the same experimental setup as in the former research [9,10] has been applied in the present work. The system studied is a planar semiconductor-gas discharge gap (SDG) device. The discharge gap of the device is bordered by a transparent anode and a Si⟨Zn⟩ high resistance electrode. The diameter of the discharge channel is 20 mm. The thickness of the gap and of the silicon electrode are 0.8 and 1.0 mm, correspondingly. The gap is filled with nitrogen. The overall structure is cooled down to the temperature $T \approx 90$ K. The spatial distribution of the light density emitted by the discharge can be observed through the transparent anode. Value of resistance of the semiconductor electrode can be controlled by illumination of the photosensitive semiconductor electrode with ir light. This permits one to vary the current, which is the control parameter of experiments described in the present work, at a constant amplitude of the feeding dc voltage.

The photoelectrical control of current, which is a local process, gives the possibility to change *in situ* the aspect ratio of the pattern forming system by a simple variation of the dimensions of the illuminated area of the silicon wafer. The brightness of the discharge in the device is proportional to the current density to a very good extent [10]. Thus, analyzing the images of the discharge, we get information about the spatial distribution of current density across the discharge area. Because both stationary and nonstationary patterns are formed in the SDG system under the variation of the control parameter, a camera with gated microchannel plate has been applied in experiments. The exposure time used was in the range 0.1–1.0 msec.

For small values of the global current the system shows a spatially homogenous distribution of the current density. The increase of the current is followed by the creation of spatial structures in the discharge gap. At a proper choice of parameters the first bifurcation observed is the transition to small amplitude either hexagon or stripe patterns [9,10]. In the present article we focus on the strongly nonlinear domain of the pattern formation process, that is, we are interested in patterns of large amplitudes which are far away from the harmonic appearance.

The characteristic stages of the evolution of the discharge in the 2D system under variation of current in a broad range are shown in Fig. 1. Starting from a low current density j we observe a homogeneous stationary state Fig. 1(a), followed by a transition state Fig. 1(b) and by a stationary stripe pattern Fig. 1(c). The transition state is not stationary, instead we observe continuous transformations of the spatial configuration of the current density distribution: The pattern may have in turn hexagonlike and stripelike appearances. This nonstationary behavior

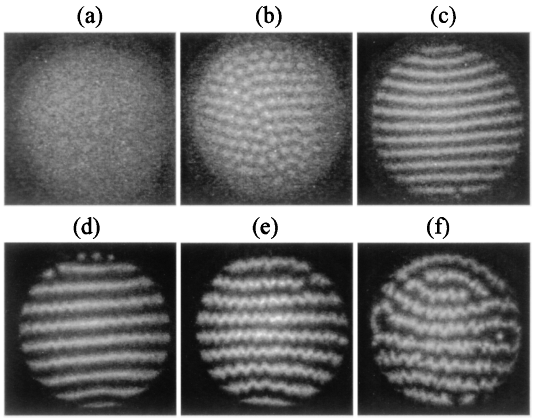

FIG. 1. Appearance of the zigzag instability for stationary stripes. From the homogeneous state (a) the system goes to the nonstationary hexagon + stripes mixed state (b), then to a pure stripe state (c), which is stationary. The further increase of current is accompanied by the growth of the spatial period of the stripe pattern and, at some critical current, by the zigzag instability of stripes (d), (e). Finally, the system goes into a complicated nonstationary behavior, a snapshot of which is shown on picture (f). Parameters: The diameter of the active area 20 mm; the width of the discharge gap 0.80 mm; the thickness of semiconductor (Si⟨Zn⟩) electrode 1.0 mm. Pressure of nitrogen in the gap: 17.4 kPa; temperature of the structure: 90 K; amplitude of dc voltage feeding the system: 2.48 kV. The current is controlled by ir light, which excites spatially homogeneously the semiconductor electrode. The current density, averaged over the area, is (a) 4.8; (b) 6.2; (c) 10.6; (d) 24.7; (e) 34.9; (f) 57.6 $\mu A/cm^2$.

has not manifested any pronounced temporal regularity. Such a turbulentlike charge transport in the electronic system which in the present work has been observed at very low current densities, seems to have the same origin as the phenomenon of "chemical turbulence" detected in planar chemical gel reactors in the range of coexistence of hexagon and stripe patterns [11].

The stripe pattern exists in a rather broad range of the control parameter $j \approx 10-25\ \mu A/cm^2$. It has been found [10] that the increase of the current in the range of the existence of a stripe pattern is accompanied by the continuous growth of its spatial period; see also Figs. 1(c), 1(d). The quantitative analysis shows that the growth of the period is accompanied by the swelling of higher harmonics in Fourier spectra. This behavior has been observed to be related with the steepening of walls of stripes. We may conclude that we enter the region of nonlinear stripes which are more and more distant from a harmonic pattern.

Finally, the next bifurcation occurs: Stripes lose their straight shape and show a pronounced zigzag structure [Figs. 1(d), 1(e)]. Note that his new instability gives a short wavelength near its threshold; see Fig. 1(d). When a large part of the stripe pattern undergoes the zigzag destabilization, the additional spatial order is expressed by the emergence of side satellites to the main maxima in the Fourier spectra. It has been noticed that the positions of these spectral components in k space (that is, the wavelength of the zigzag) do not depend noticeably on the value of the control parameter. The observations show also that the zigzag structure is not stationary: While making acquisition of patterns with the highest repetition rate available with our equipment (which is 10/sec) we have observed different spatial realizations of the zigzag deformation of a stripe in subsequent frames. So the instability creates the wave which moves along a stripe.

While driving the 2D system into the domain where stripes are formed, we inevitably have to do with the situation of multiple stripes. The question arises of whether or not the observed phenomenon of zigzag destabilization is the feature of a solitary stripe, or it is specific only for the ensemble of them? To answer it, experiments on a system with a low aspect ratio along one (lateral) direction have been made: Instead of the homogeneous illumination of the total aperture, only a zone along its diameter is activated with the light. Thus, the active area of the experimental system is now of rectangular shape. The short dimension of the illuminated area has been about 2.0 mm, which is on the order of one period of the stripe pattern in the 2D system.

Now, the first bifurcation from a structureless state of Fig. 2(a), observed at low currents, is followed by the formation of a periodic 1D pattern, Fig. 2(b). Evidently, this pattern can be considered as the analog of the state, shown in Fig. 1(b) for the 2D domain. The next bifurcation creates only one stripe, Fig. 2(c). We shall stress that this state has to be referred to as the localized stripe, generated by the constricted system: It is characterized by the narrowing of the transversal spatial distribution of brightness as compared with the corresponding distribution in a stable domain, before the first bifurcation has occurred; see Fig. 3. At some stage of the system's evolution the next bifurcation occurs: The straight stripe undergoes the zigzag destabilization; see Figs. 2(d), 2(e). Experimental observations suggest that

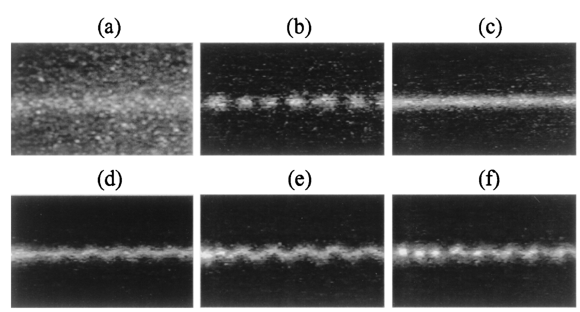

FIG. 2. Development of the pattern in the constricted system. The experimental parameters are the same as for the data of Fig. 1, but now only a strip zone of the width of ≈ 2 mm of the semiconductor electrode is activated with IR light. Dimensions of the shown fragment, $H \times V = 11 \times 6.7\ mm^2$. Global current: (a) 9.5; (b) 12.7; (c) 16.2; (d) 31.4; (e) 40.7; (f) 49.1 μA.

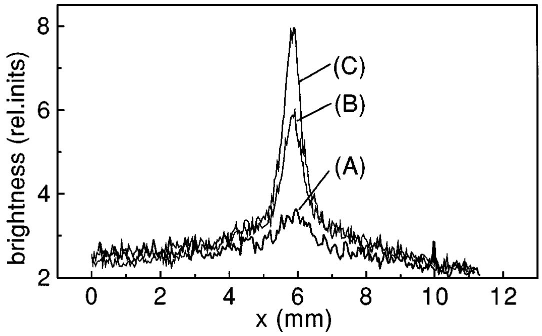

FIG. 3. Transversal profiles of the spatial distribution of the discharge brightness in the quasi-2D (constrained) system under the current increase. For the experimental conditions, see the caption of Fig. 2. The curve (*A*) refers to the state of the system before the first bifurcation to the periodic pattern (b) of the Fig. 2 has occurred. Curves (*B*) and (*C*) are obtained in the range of existence of the solitary stripe [see the pattern in Fig. 2(c)]. Global current: (*A*) 9.5; (*B*) 16.2; (*C*) 23.4 μA.

this bifurcation proceeds supercritically: The continuous growth of the amplitude of the zigzag is observed as the driving current increases after passing some threshold value. The zigzag deformation of a solitary stripe is not stationary and is expressed as a traveling wave. As in the experiments on the 2D domain, our equipment could not provide sufficient temporal resolution to evaluate the velocity of waves propagating along a stripe. The state with the zigzag stripe has been revealed to exist in a rather broad range of current. This has permitted one to evaluate the dependence of the spatial period of the zigzag on the current; see Fig. 4. Analogously to the 2D case, the spatial period is only slightly dependent on the current, its value being close to that observed on the 2D system.

It is remarkable that at higher current the next bifurcation is observed: The tendency of breaking a zigzag stripe into spots can be seen in Figs. 2(e), 2(f). On this stage the overall pattern shrinks in the transversal direction, Fig. 2(f). The observed phenomenon can be referred to as the fragmentation of the LS. We shall stress that a similar effect has been noticed also in the 2D case, in the course

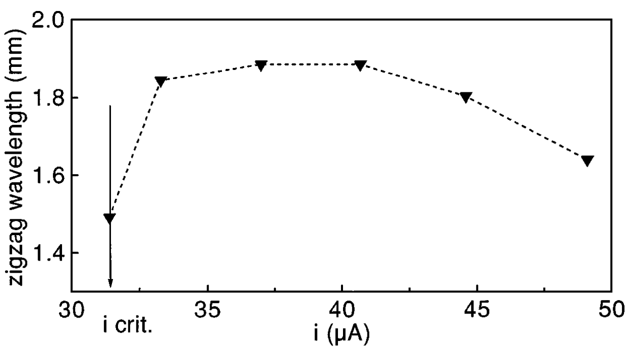

FIG. 4. Period of the zigzag traveling wave for a solitary stripe versus the global current of the device. For the experimental conditions, see captions of Figs. 1 and 2. *i crit*: Critical current for the zigzag destabilization.

of the continuation of the scenario shown in the Fig. 1, when increasing the current further. The distribution of the brightness along individual stripes becomes spatially modulated, and their fragmentation proceeds. During their movement fragments of a broken stripe can combine with products of the decomposition of other stripes in the system. The described process of stripe fragmentation and the following reconstruction of the pattern observed on the 2D domain, seems to be chaotic in time.

Summarizing the experimental results, we can state the observation of analogous scenarios both for 2D system and for the constricted (quasi-2D) domain. The first bifurcation in both cases is accompanied by the creation of low amplitude patterns. Patterns in current density distributions of dc driven extended SDG structures have been shown [12] to form due to the competition between the *autocatalytic* processes in the current transport in the discharge domain, and the *inhibiting* role of spatially distributed load to the discharge which is provided by the resistive semiconductor electrode. The inhibition proceeds at the expense of the lowering of electric potential in domains of increased density of current. For spatially nonhomogenous perturbations the characteristic length of the lateral spreading of the potential normally exceeds that for the charge carriers inside the discharge gap. In activator-inhibitor terms this means that the range of spatial spreading of the autocatalytic variable (density of charged particles in the gap) l is shorter as compared with the corresponding value for the inhibitor (electric potential) L, and the conditions of the Turing-like destabilization of the considered electronic system with lateral inhibition can be met. So, small amplitude patterns of Figs. 1(b), 2(b) can be referred to as Turing-like structures [9,10].

It is essential that the SDG system with the cryogenic discharge shows these primary bifurcations at low pumping current, which is only $\sim 5\ \mu\text{A/cm}^2$. Present experiments demonstrate that a sequence of next bifurcations can be detected while increasing the current further. Large amplitude stripes in the spatial distribution of electric current may have grown and at some critical current undergo the zigzag instability. According to experimental results, this instability proceeds both for solitary stripes, and for an ensemble of them. Because the main regularities of the observed phenomena are the same in both cases, we can state that the effect is specific for solitary stripes. It is important to stress this point because the phenomenon of zigzag instability for periodic roll/stripe patterns has been observed and well understood both for hydrodynamical [13] and R-D [3] systems, where it can be treated in the frame of weakly nonlinear analysis.

The problem of stability of localized stripes in R-D systems has been theoretically treated in [4–8]. Bifurcations not only to stationary zigzag modes, but also to traveling wave solutions have been revealed [6,7]. Experimental data of our work provide a general confirmation of these theoretical findings related to the existence both of zigzag instability of solitary stripes itself, and of the traveling zigzag mode. At the present stage of the research we are not able to give a concrete comparison of theory and experiments, because the detailed physical model of processes in the experimental system is still not available. We note, however, that in the experiments described in the present work, the zigzag destabilization is observed for narrow (spike) stripes, see Fig. 3, whereas the theoretical models [6,7] deal with so-called "broad" [5] stripes. These are stripes for which the width of walls is much less than the width of the flat band domain, which is filled with the increased concentration of the activator. For some models of R-D processes analyzed in Chap. 13 of Ref. [5], the critical wave vector k_c of the transversal destabilization of a solitary stripe is expressed as a function of the product of the characteristic lengths of diffusion for activator and inhibitor: $k_c \propto (lL)^{-1/2}$.

This means that the zigzag deformation of a stripe may have its own characteristic wavelength, which is controlled by the local diffusion interaction of activator and inhibitor, and is not essentially determined by, say, boundary conditions. This result can be interpreted in the sense that the destabilization of stripes may occur via a Turing-like scenario. The wavelength of such a zigzag structure should be of the value close to that of a primary, small amplitude Turing pattern which can exist in the system. The pictures of Figs. 1(b), 1(e) for the 2D domain and of Figs. 2(b), 2(e) for the quasi-2D case which contain both small-amplitude primary patterns and developed stripes destabilized to zigzag, just exemplify such a regularity.

Finally, the intriguing question arises of whether or not the observed fragmentation (breaking) of stripes at large currents is due to the intervening into a domain of parameters, where they become destabilized both to zigzag and varicose modes, simultaneously. This possibility has been considered in [6,7]. The expressed irregular spatiotemporal behavior observed in the 2D domain [the beginning of this process is presented by snapshot (f) of Fig. 1] may provide an example of the turbulency in the ensemble of localized states of the active medium when conditions for both instabilities are fulfilled.

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