Zigzag Destabilized Spirals and Targets

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We report experimental evidence of novel kinds of patterns—zigzag destabilized spirals and targets. They have been found in a planar dc driven semiconductor–gas discharge system. Comparison of these patterns with conventional spirals and targets that can form in the same system allows one to conclude that the observed phenomena are specific for large amplitude structures of pattern forming media. [S0031-9007(98)06511-9]

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Spirals and targets are among the most interesting spatiotemporal patterns which exist in nonequilibrium systems. They are observed in systems whose internal structure and underlying processes, which determine their dynamical properties, are quite different: Examples are classical Belousov-Zhabotinsky reactors [1,2], chemical reactions on surfaces [3], systems with Rayleigh–Bénard convection [4], semiconductors [5], and populations of microorganisms [6]. It is believed that spirals are important in controlling rhythms of the heart activity in highest organisms, including humans [7].

The question arises of whether spiral and target patterns in dynamical systems can undergo transitions to patterns of the next level of complexity, while retaining their global shape. For example, could there exist spirals with a substructure? While the study of spirals and targets has been done for a long time, we are not aware of any experiments giving a positive answer to this question.

In the present Letter we report on the experimental observation of spirals and targets whose arms and rings, respectively, are zigzag destabilized. Experiments have been done on an electronic system which is a dc driven planar semiconductor–gas discharge (SGD) device [8–11]. Patterns in the system are formed due to the coupling of processes of charge transport in two layers, one being a linear high resistance semiconductor, while the other one, a gas discharge domain, is a medium the transport properties of which are strongly nonlinear; see Refs. [12,13]. Patterns in the device can easily be observed by studying the distribution of the discharge glow in the gap. The data can be used for a quantitative evaluation of emerging patterns [10].

In the current work silicon doped with deep impurities of Zn or Pt has been used as a high resistance semiconductor electrode. Wafers of a diameter of 30 mm and of a thickness of $d_s = 1$ mm have been applied. The effects observed do not depend on the kind of semiconductor material. The discharge gap has a width d_g on the order of 1 mm and is filled with nitrogen at the pressure p. Observations of spatiotemporal structures in the device have been done in the range of $d_g = 0.55–1.4$ mm. To provide a high initial resistance state of the semiconductor the device is cooled down to $T \approx 90$ K with liquid nitrogen. The resistivity ρ of the semiconductor electrode can be controlled by infrared (ir) light which generates nonequilibrium charge carriers in the silicon crystal. The entire system is fed with dc voltage. Both the light intensity and the feeding voltage can be used as convenient and independent control parameters.

The considered system manifests formation of both (quasi)harmonic small amplitude structures and patterns containing large amplitude elements [localized states (LS)] in one and the same setup, while only control parameters are changed; see [9]. A complete description of the system's behavior in the parameter space has not been made so far. We may state, however, that at high values of ρ in general harmonic patterns can be observed [8,10], while a decrease in ρ is accompanied by formation of LS's; see [9,11]. In the present study we deal with both modes of pattern formation. Additional details to the experimental setup can be found in the above cited Refs. [8–11].

Figure 1 shows the global current-voltage characteristics of the device obtained for two values of the intensity of ir light that controls the resistivity of the semiconductor electrode. For voltages that are higher than some critical value, the discharge is ignited, and an electric current can be detected. At a further voltage increase a continuous growth of the current is observed. Related distributions of the discharge glow are depicted in Fig. 2.

Increasing the feeding voltage for high resistivity ρ of the semiconductor electrode, we observe the following sequence of patterns: hexagons → stripes → stripes with defects → patterns containing rotating spiral waves [Figs. 2(a)–2(e)]. Patterns with spirals seem to show in general a nonregular spatiotemporal behavior. This scenario is similar to that observed in certain problems of Rayleigh–Bénard convection when changing the temperature difference across a layer of a studied sample [14]. It is known that spirals in convection flows can give rise to chaotic behavior of a system, so-called spiral-defect chaos; see [4] and one of the recent papers on this subject [15]. Transitions from hexagons to stripe patterns and further to nonstationary patterns have been also found in chemical reactors, which are reaction-diffusion systems;

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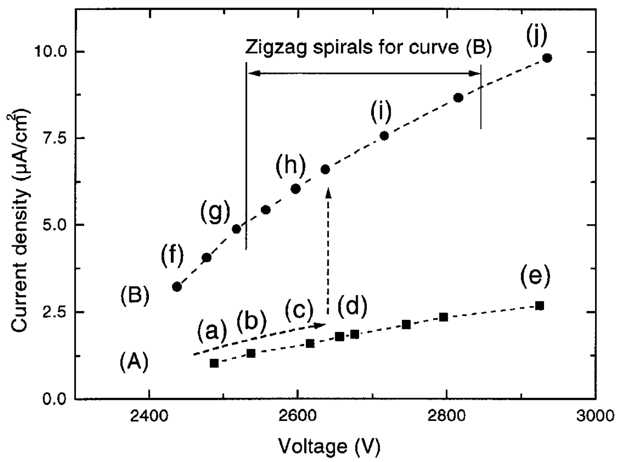

FIG. 1. Current-voltage characteristics of a planar semiconductor–gas discharge device for two resistivities of the semiconductor electrode, $\rho = 1.6 \times 10^9\ \Omega$ cm, (*A*) and $3.5 \times 10^8\ \Omega$ cm, (*B*). The current density is averaged in space and time over the active area of the system. The ρ value is controlled by ir light due to the photoelectric effect in the semiconductor electrode. Labels on the curves mark domains where corresponding patterns given by Fig. 2, are observed in the device. The dashed arrows show a route to obtain zigzag spirals via the stage of conventional spirals. The structure is driven by dc voltage with the negative polarity at the semiconductor electrode. The discharge gap is filled with nitrogen. Parameters: lateral extension of the discharge area is 20 mm; $d_g = 0.8$ mm; $d_s = 1.0$ mm; N_2 pressure $p = 1.5 \times 10^4$ Pa; $T \approx 90$ K.

see Refs. [16]. So, Figs. 2(a)–2(e) demonstrate clearly that extended electronic media can also exhibit typical scenarios in development of harmonic patterns known from the study of hydrodynamical, chemical, and other systems. We remark that bifurcations which include transitions to hexagon, stripe, and (stripe + defects) patterns, can be analyzed in the frame of the weakly nonlinear analysis, which includes the interaction of a small number of harmonic modes; see [14,17]. We therefore conclude that in Figs. 2(a)–2(e) we deal with low amplitude structures.

A decrease in ρ of the semiconductor electrode has a drastic influence on the pattern formation process; see Figs. 2(f)–2(j). While the definite similarity exists in both scenarios, now patterns acquire a new quality. The "hexagon" stage of the system [Fig. 2(f)] is characterized by the existence of bright spots in the discharge glow. This state is essentially nonstationary and spatially disordered. Spots move across the active area of the experimental cell. The number of spots varies: Some spots collide and fuse, while new ones are generated at the periphery of the system. Occasionally, spots organize a well-defined hexagonal arrangement, as shown in Fig. 2(f). One can say that in this case the pattern has lost its harmonic appearance and represents an ensemble of LS's with their specific quasiparticle features [11,18].

It is also remarkable that in general the observed spots have an internal structure. This is expressed via their breathing dynamics which may be accompanied by the loss of the radial symmetry. In the latter case the transition to *starlike* spots takes place. These phenomena evidently represent secondary bifurcations of solitary spots and are interesting enough to be studied separately. Here, we notice only that on the breathing regime of a chemical reaction it has been reported in Ref. [19]; breathing localized spots have been recently observed in a planar ac driven gas discharge system [20]. In our case the speed of internal dynamics of solitary spots is rather high. The available rate of the image acquisition equipment applied in the present research (which was 8 Hz) is not high enough to follow the real internal dynamics of spots. In order to stress the difference between spots which are stable and thus can build up a stable structure, and the present situation where spots undertake further bifurcations, we will refer to the latter objects as *large amplitude spots.*

Now, starting from a pattern in the parameter range of large amplitude spots [Fig. 2(f)], an increase in voltage is accompanied by the creation of spatially extended constituents, which are built up of spots that stick together;

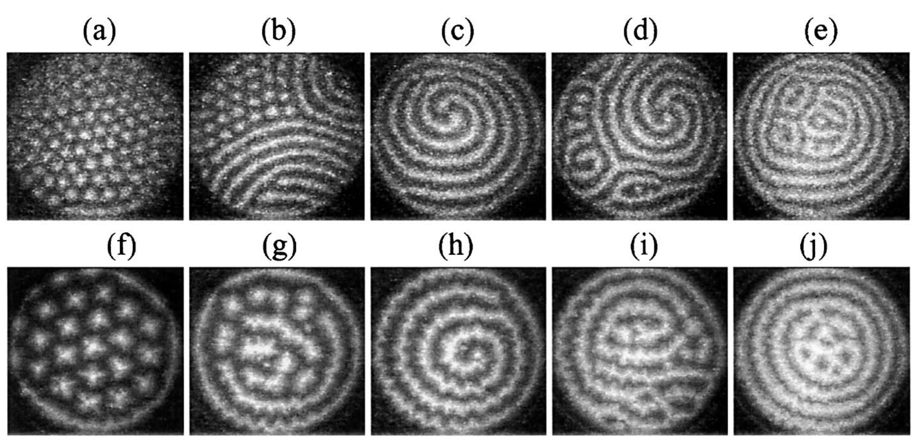

FIG. 2. Characteristic bifurcation scenarios for two values of resistivity of the semiconductor electrode. The sequence of snapshots (a)–(e) and (f)–(j) have been obtained while going along curves (*A*) and (*B*) in Fig. 1, respectively, by increasing the voltage. An intensified gated CCD camera was used to capture images of the discharge glow. The exposure time lies in the range 0.3–1.0 msec.

see Figs. 2(g) and 2(h). Thus, new elements of the total pattern are organized by two, three, or more large amplitude spots. These extended elements have a stripelike appearance; see Fig. 2(g). They fit together with a tendency to generate a zigzag structure which primary "building" elements are breathing spots. The state of the system remains nonstationary. Stripes move over the active area and may decompose into smaller fragments, while new extended constituents form via accumulating spots that are available. At further voltage increase these *large amplitude stripes* invade all the active space, and at some stage spirals with a spatial zigzag modulation along their arms are organized; Fig. 2(h).

Zigzag spirals exist in a broad range of the driving voltage; see Fig. 1, curve (*B*). In the course of time spirals are destroyed and again generated. Their destruction may proceed via the decomposition into smaller fragments of a pattern. These fragments interact with each other, so that an apparently spatiotemporal chaotic behavior establishes. Such a temporally disordered mode of the current transport again can be suppressed later, due to the appearance of a new extended spiral (that is, of a spiral which occupies practically all the area of the system). Before being destroyed, a spiral can make a number of rotations. The lifetime of a spiral normally does not exceed some seconds. During its life a spiral exemplifies a complicated trajectory of the tip similar to what is observed in chemical systems (see, e.g., Ref. [2]). The characteristic rotation frequency of spirals is 10–15 rad/sec for present experimental conditions. A spiral may undertake also the transition to a target. Such a process can be seen in Fig. 3 where an example of the temporal evolution of a pattern at constant values of control parameters is given. This scenario includes transitions from a spiral to a target and vice versa. Figures 3(a)–3(d) explicitly show how in the course of these transitions the direction of a spiral's rotation may change.

In the present experiments we have two independent control parameters, the feeding voltage and the intensity of ir light. So, one and the same final state can be reached by a number of procedures. For example, one can create zigzag spirals by going along the route indicated by Fig. 1 by dashed lines with arrows. Such a scenario contains a transition from small to large amplitude spirals. We remark, however, that this transition is not sharply defined: In effect, this is a continuous transition from a state where conventional spirals dominate, to a state with zigzag spirals.

The quantitative difference between conventional and zigzag destabilized spirals is observed on spatial profiles of electric current density for these patterns; see Fig. 4. We see that for a zigzag destabilized spiral the amplitude and the spatial period of the pattern, as well as characteristic spatial gradients of the current density, increase as compared to a conventional spiral [cf. curves (*A*) and (*B*)].

While the data presented above refer to the fixed value of the width of the discharge gap $d_g = 0.8$ mm, formation of zigzag destabilized spirals and targets has been observed in the whole range of d_g, from 0.55 to 1.4 mm, studied in the current work. When increasing (decreasing) d_g, one should generally decrease (increase) the nitrogen pressure in the gap. The robustness of the effect with respect to variations of the interelectrode distance of the discharge is evidently related with the invariance of properties of low current discharges, when keeping constant the product $p \times d_g$; see [21].

Though there have been reported so far no theoretical results which would correspond to the observed phenomena of zigzag destabilization of spirals and targets, it is hoped that these effects can be understood on a phenomenological basis. It was shown earlier that the underlying mechanism for pattern formation in semiconductor–gas discharge systems can be attributed to the autocatalytic behavior of current transport in the discharge domain and to the inhibiting voltage drop at the distributed resistive electrode. For a spatially extended system two component reaction-diffusion models can be applied for the analysis of pattern formation [11–13].

As argued above, zigzag spirals are observed in *large amplitude* patterns. It is appropriate to notice that earlier the phenomenon of the zigzag destabilization of solitary stripes (which are examples of LS's) has been observed in the same system [9]. This effect appears when the amplitude of a stripe reaches some critical value. Theoretically, secondary bifurcations of LS's in reaction-diffusion systems, including the zigzag instability of solitary stripes,

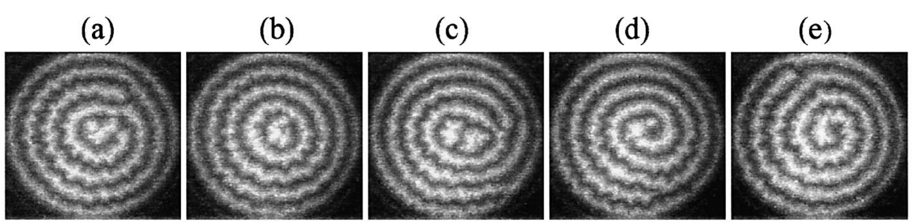

FIG. 3. An example of transitions *spiral* → *target* → *spiral*. The total duration of the recorded scenario is about 10 sec, the exposure time to take one picture is 0.4 msec. Time increases from left to right. In the course of time the direction of rotation of the spiral changes [cf. pictures (a), (d), and (e)]. The time interval between snapshots (a),(b), and between (c),(d) is 125 msec. This behavior has been recorded at constant values of control parameters, in the domain (h) of the current-voltage characteristic (*B*) of Fig. 1.

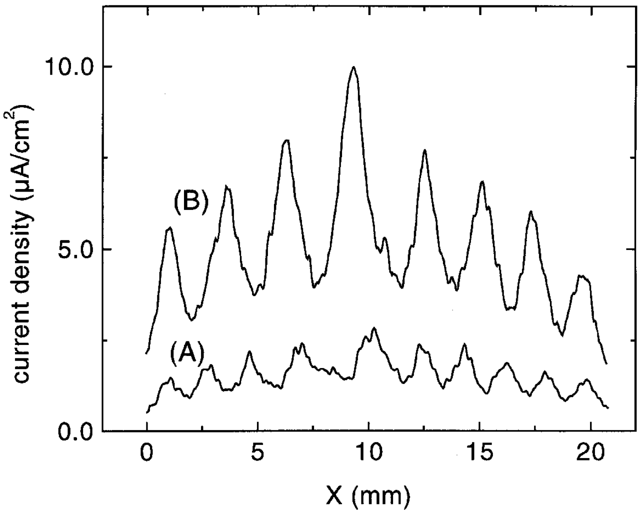

FIG. 4. Spatial profiles of the conventional (*A*) and a zigzag destabilized spiral (*B*). The data refer to central domains of images (c) and (h) of Fig. 2, respectively. To obtain the curves, spatial distributions of the discharge brightness have been rescaled to current density profiles using the calibration routine similar to that applied in Ref. [10].

have been discussed in Refs. [22]. We suppose that by observing a transition from a low amplitude to a large amplitude spiral (seen in the current density profiles of Fig. 4) we witness the transition to a solitarylike object in the sense that arms of a large amplitude spiral acquire the properties of a localized (*large amplitude*) stripe. The growth of the amplitude of patterns seems to provoke their zigzag destabilization.

In conclusion, we have shown that by variation of control parameters the mode of formation of spatiotemporal structures in a system can be changed: From low amplitude (harmonic) patterns there can be made a transition to the regime of large amplitude localized states. New patterns—zigzag destabilized spirals and targets have been found under these conditions. As dependent from a route of variation of control parameters, zigzag destabilized patterns can be obtained either from low amplitude patterns, or can be assembled by large amplitude spots which are unstable and show breathing dynamics. This scenario has some analogy with the known and well-understood transition between low amplitude hexagonal and stripe patterns, but proceeds in the domain of parameters, where on both sides of the transition patterns are far away from a harmonic appearance. Experimental data of the work refer to the system which can be considered as a variant of reaction-diffusion systems. The analogous phenomena should exist in systems of a different nature which can organize localized states.

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